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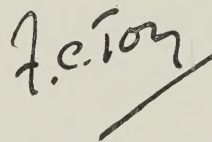
## Foreword

THE application of physics in industry has been one of the particular concerns of the Institute of Physics since its foundation in 1918. As early as 1923 it commenced the publication of a series of monographs and lectures under the general title of *Physics in Industry*. Although it has not hitherto been possible to establish a Journal devoted to applied physics generally, the Institute's *Journal of Scientific Instruments* has carried for the past fifteen years a number of contributions of this nature, and for the past two years these have been gathered together in a separate Section and the title enlarged to *Journal of Scientific Instruments and of Physics in Industry*. Fission—to use a current term—has now taken place, and the “Physics in Industry” section forms the basis of this new *Journal*, whilst the older *Journal* will again be devoted solely to scientific instruments and apparatus. For these reasons the format has been chosen to correspond with that of the original *Journal*.

The primary object of the *British Journal of Applied Physics* is to announce new applications of physics and developments of those already known. This will be achieved by accepting original papers of scientific merit, by publishing special articles surveying particular aspects of a subject and also a selection from the lectures, discussions and symposia arranged by the Institute and its Groups and Branches at home and overseas. Sections will also be devoted to book reviews, correspondence and technical notes and news.

At the Royal Society's Scientific Information Conference last year the need for a great improvement in the presentation of scientific papers was repeatedly stressed. In response to this, every endeavour will be made to establish and maintain a high standard in this respect.

We request our many friends all over the world to support this *Journal* by contributing to its pages, by subscribing to it, and by encouraging others to do so also. We are very grateful for the help we have received from the Council of the Royal Society in the form of a grant towards the cost of this new venture, and we also record here with pleasure the sympathetic support and encouragement we have received from the Council of the Physical Society.



President,  
*The Institute of Physics.*



## SPECIAL ARTICLES

## A Scientific Education\*

By SIR PHILIP MORRIS, C.B.E., M.A., Vice-Chancellor, University of Bristol

The rapid development of science raises so many immediate problems that we can hardly find time to realize what a revolution it has made in all our thinking and behaviour. The immediate problems have been partly solved by including science in the educational curriculum, but science is superimposed on a system which is essentially unchanged. Scientific training is distinguished from liberal education and regarded as an encroachment upon these humane studies, which it threatens to displace. Yet, whatever the subject matter, the knowing mind observes the same principles and limitations as far as its own processes are concerned. Investigation into the diverse needs of students, as well as the demands of the subject, should discover principles and methods for making the study of science a satisfactory education as well as an adequate training.

The importance of Science in the modern world is too often seen in too narrow a perspective. We concern ourselves with the importance of scientific research, with avenues of employment for scientists, with the relationship between scientists and administrators or policy-makers, and many such questions which are incidental to the problems of the times. The direct bearing of scientific discovery and scientific training upon national defence at a time when, in the world generally, the foundations of peace are insecure tends to make us concentrate so much upon the urgent problems of the moment that we have too little time to see the development of science in a wider perspective and from a long-term standpoint. As it seems to me, we are living at an advanced stage of a movement in human affairs which can reasonably be called a scientific revolution. It is a movement which in all probability has not yet reached its peak, although it has already immeasurably quickened the movement of affairs. In some directions the scientific discovery and advance has wrought more changes more quickly than in others. For many years past this has been the case, and for many years to come it will probably remain true, so that there has not been a natural opportunity to take stock of the situation and to see how far the centre of gravity in intellectual affairs has been moved and is still moving. It seems to me, and no doubt also to many of you, that it is high time that we made for ourselves an opportunity to take stock of the state of affairs and by discussion and thought tried to offset the general intellectual disequilibrium which has resulted from the unequal rate of change.

For more than a century now scientific discovery and advance has, more than any other single cause, influenced the great changes which have come about in social, economic and intellectual affairs. In social affairs it has

made possible—it may almost be said to have caused—a radical change in the size, composition and distribution of the population. During this period of scientific quickening the population of our own country has increased almost fivefold and a community which was capable of being self-dependent and largely agricultural has become preponderantly industrial and very largely dependent upon the productivity of other parts of the world. The kind of activities which engage the attention of the majority of the population for the greater part of their time have now become activities which rely upon the scientific progress of the past century and involve a degree of scientific understanding, even if it is largely unconscious, which was unknown to our grandfathers. Natural forces which to our grandfathers were still matters of superstition and poetic fantasies have become for us and our children common and everyday means of doing common and everyday things. Not only has the whole pattern of social life changed, but because of it and simultaneously with it the spontaneous patterns of thought have changed also. The metaphors and imaginative analogies of yesterday have in many cases become discredited because they seem to imply an altogether false interpretation of the nature of the physical universe and they have been very largely replaced in common parlance by an inaccurate use of scientific terminology and analogy. Metaphors and parables drawn largely from natural sources which for centuries did service to bring home to successive generations persisting truths are less and less serviceable. This has not only been a loss to literature and religious exposition, but something more serious still has occurred. It has too frequently and too easily been assumed that the discrediting of the metaphors and analogies has also discredited the truths which they had been used to express and communicate.

The economic developments occasioned by scientific advance have been no less rapid than the social. At this particular period of world-wide economic dislocation this aspect of the subject we are considering needs

\* Based on the opening of a discussion on the subject at the inaugural meeting of the Education Group of the Institute of Physics held on 5 October, 1949, in London. Membership of the Group is open to all interested; further details may be obtained from the Secretary, the Institute of Physics, 47 Belgrave Square, London, S.W.1.



no emphasis. Side by side with these economic developments and greatly influenced by two world wars, there have grown up political problems of great difficulty and complexity. It is a commonplace that while the map of the economic and intellectual world has been changed beyond recognition, the development of political and economic concepts has been much slower, so that our intellectual experience and resources seem to be inadequate to the social and political problems which scientific and mechanical advances have caused and will continue to aggravate. The changes in the map of the intellectual world have been accompanied by a changed intellectual outlook. New scientific views "by an astonishing succession of intellectual and experimental achievements have largely and for ever removed former acceptations of human thought. The mental habits of two thousand years and more are overturned by several propositions. . . ." The majority of and the most important problems, both domestic and international, are unmistakably complex and demand for their fair consideration at least an appreciation of all the factors involved, many of which, and by no means the least important, are scientific. Such an appreciation demands to a rapidly increasing extent in all concerned some acquaintanceship with the ever-growing area of the intellectual map which science has charted.

If we look at the development of our educational curriculum we see that much of the perplexity through which Man has been living during the last century is reflected in the content of the education of children and young people. The nineteenth century saw battle royal done for the addition of scientific subjects to the traditional educational curriculum which had come down to us, modified and transmuted but not radically changed, through the Middle Ages. The battle for the inclusion of scientific subjects was not solely an intellectual fight. Huxley's *Essays on Science and Education* epitomized the extent to which the invasion of the curriculum by scientific subjects was seen to be a challenge to much that was traditionally sacred, and important cross-currents in the battle were concerned with general questions of intolerance and emancipation which were already acute on other grounds. The right of one scientific subject after another to a place in the curriculum was, it is not untrue to say, grudgingly conceded and its presence was to a greater or less extent tolerated rather than welcomed. The reasons for this can with a little imagination be readily appreciated and understood, but merely to understand the reasons is not enough. The result was that the scientific subjects became an accretion upon an existing curriculum, were not assimilated to it and tended to remain what might in the political field be called an "alternative form of intellectual government." In the early stages those who took up scientific subjects and who made the cause of science in the curriculum their own, had been powerfully influenced by the traditional curriculum which had played a large and important part in their early education. The in-

fluence of the traditional curriculum upon scientists had been diminishing and is still diminishing so rapidly that we are approaching the point at which those who are to achieve any real standard in a scientific subject must find the whole of their education in their scientific pursuits. The question, therefore, that we have to ask ourselves is whether the scientific subjects in the curriculum are to be regarded as the vehicle of a complete education or as a means merely of scientific training.

The development of thought about the curriculum, or what might be called the philosophy of the curriculum, has proceeded extremely slowly, while the curriculum itself has changed with great rapidity. If the curriculum of a grammar school to-day is compared with a typical curriculum of a grammar school of, say, only seventy-five years ago, the obvious differences are so great that one would expect to find at least a considerable change in the explicit philosophy of the curriculum. It is probably true, however, that the philosophy of the curriculum has had to take care of itself while the curriculum itself, under other influences, was undergoing radical changes and consequently the philosophy of the curriculum has not developed correspondingly.

It is impossible to develop this argument far. It must suffice to ask whether the philosophy of the curriculum implicit and, indeed, explicit in Newman's *Idea of a University* can be matched by any more recent statement of it in terms of the present state of affairs. Newman wrote at a time when it was quite compatible with the normal acceptations of thought that there was a hierarchy of subjects, some of them being more inherently significant and important than others, some of them being liberal and others of them "illiberal." This attitude towards the subjects was traditional and it is probably true that, while it finds less favour nowadays, it still influences much of our thought and discussion. We have to contrast a statement that subjects dealing with human values are necessarily liberal with the claim made in some quarters that scientific subjects must be leavened by subjects dealing with human values if a scientific education is also to be a liberal education. The way in which we at present think about these things is to be inferred from the way in which we talk about them. It is almost a common acceptance of conversation to refer to "a classical education and a scientific training." There is implicit in this way of talking the view that scientific subjects are suitable means of training scientists, but not by themselves a suitable vehicle for the education of men and women. There has been much philosophical thought which has been more progressive and we shall all remember such statements as "No subject is essential to a liberal education," which carries the obvious implication that there is no natural or necessary hierarchy in the subjects into which knowledge is divided and which indirectly suggests that the educational value of the subjects of knowledge depends upon their use and treatment rather than upon their matter. Therefore it is perhaps pertinent for me to ask



whether what passes for an adequate scientific training necessarily has as a by-product a satisfactory education, or whether it is necessary to consider much more fundamentally than has so far been done how scientific subject matter at the elementary stages should be arranged and utilized in order to give not only a foundation of scientific knowledge and technique, but also a sound general education.

I mentioned earlier the way in which we customarily talk of "a classical education and a scientific training" and the way that we have got into of assuming that there is little training in the former and still less education in the latter. Believing as I do that scientific subjects can be the means to a liberal education if wisely and properly used to that end, I believe it is time that more serious consideration was given to the effective employment of scientific subjects for the purpose of the education of men and women. There are formidable difficulties in the way, but any satisfactory reform of the curriculum depends upon a belief that obstacles, though formidable, are not insuperable. It is my hope, therefore, that the Education Group of the Institute of Physics may be able to make some effective progress in this direction. The difficulties in the way are all too familiar. What used to be called Natural Philosophy, and later Natural Science, has, as a result of very large and rapid developments within comparatively narrow fields, become broken into many fragments and the fragments themselves are jostling with each other to find their proper relationship in a systematic organization of knowledge. Scientists generally have become more and more experts in a scientific subject and are themselves vocal about their incapacity to deal with subject matter which lies outside their immediate field. New interpretations of natural phenomena have done much to upset the more elementary stages of scientific subjects so that the method and order of developing the scientific subjects in schools both tend to follow too closely the division into subjects and branches found in the higher stages of scientific work and also to be upset by scientific development and discovery. At the undergraduate stage the quantity of matter held to be necessary in any of the scientific subjects and the amount of time thought to be essential to the development of the appropriate technique have now become excessive and unduly oppressive. Further, the degree of versatility which we recognize in our fathers is much more difficult for our sons. I would venture even to suggest that what are held to be the necessities for the adequate development of the subject have passed the bounds which make it possible also to remember and provide for the growth and education of the student. In all educational matters consideration must be given at one and the same time to what the subject inherently requires and to what the student has the human capacity and ability to give. The over-emphasis of the former would seem to be likely to produce well-trained men and women whose education was deficient, while undue emphasis of the latter could

—and no doubt sometimes does—result in nice men and women who are prepared to use conjectures as facts and who lack the discipline of a well-established technique. Scientists have perhaps, in the heat of battle for the recognition of their subjects, too easily assumed and accepted that there are methods of thought and enquiry which are inherently scientific. They have perhaps mistaken an essential difference of subject matter and an essential difference in the possibility of verification and experimentation because of the difference of the subject matter for a different intellectual process and approach. If it could be agreed amongst scientists that, whatever the subject matter, the knowing mind observes the same principles and limitations so far as its own processes are concerned, much would be done to improve the present situation.

In the age in which we live an immense variety of boys and girls are exposed to a greater or less degree to teaching in scientific subjects. A similar variety, though not quite so extensive, is to be found at the undergraduate stage. If it be true that education must always take account at one and the same time of the needs of the subject and of the capacities and kind of interest of the student, then one would expect or at any rate hope to find some variety in method. For some students, as, for instance, the intending doctor, who needs some knowledge of physics but is not going to be a physicist, the general lines of the course should perhaps be differentiated from those of the course for intending physicists, or, again, from those of the course for intending chemists or biologists. Experience of good medical students doing post-graduate courses in radiology, for instance, seems to suggest that their early training in physics has not left much either in the way of technique or of foundation in the subject. In grammar schools a varied approach is perhaps even more necessary if all boys and girls are to have some reasonable understanding of the prevailing interpretation of the physical universe. In order to make my meaning clear I venture to suggest three differentiated methods of approach which may suit different people, or the same person at different stages in his education. There is, first, the as yet little used method which I call the "project" method and by which I mean the selection of certain particular activities within the pupil's experience involving a considerable amount of knowledge of physics and the teaching of such physics as is immediately necessary to some understanding of those activities. The second method I would call the "descriptive" method, meaning thereby the unfolding by narration and description of the development of physical knowledge so that it may be comprehended as a series of human conquests and achievements, together making a coherent story of the development of physical knowledge. The last I call the "systematic" method, meaning thereby the logical development of principles and phenomena in the manner which will most efficiently lay the foundations for serious study of the advanced branches of the subject.



Perhaps all of these methods and many others could be considered with a view to approaching the central problem which I have already outlined of ensuring that scientific subjects are so employed as to give not only scientific training, but also a general education which will include some insight into the whole range of human affairs, human nature and human purpose.

I have spoken of the importance of variety of method and treatment in the development of scientific subjects to students. It would not only be churlish, but would also be missing an opportunity, if I did not remind myself at the same time that great scientists have thought of this and expressed it in words far better than I can command. In this connexion James Clerk-Maxwell comes to my mind and he has said:—

“There are . . . some minds which can go on contemplating with satisfaction pure quantities presented to the eye by symbols, and to the mind in a form none but mathematicians can conceive.

There are others who feel more enjoyment in following geometrical forms which they draw on paper, or build up in the empty space before them.

Others, again, are not content unless they can project their whole physical energies into the scene which they conjure up. They learn at what rate the planets rush through space, and they experience a delightful feeling of exhilaration. They calculate the forces with which the heavenly bodies pull on one another, and they feel their own muscles straining with the effort.

To such men momentum, energy, mass are not mere abstract expressions of the results of scientific enquiry. They are words of power, which stir their souls like the memories of childhood.

For the sake of persons of these different types, scientific truth should be presented in different forms, and should be regarded as equally scientific, whether it appears in the robust form and the vivid colouring of a physical illustration, or in the tenuity and paleness of a symbolical expression.”

The truth is, as it seems to me, that we are at the height of a rapidly unfolding movement in human affairs, which is as significant and important as that great movement called the Renaissance which has already become historical. It has been said that “The Renaissance is the name of a European movement so gradual, broad, manifold and subtle that any attempt to reduce it to a single expression is predestined to failure” (Walter Raleigh). We ourselves are living at the height of a movement as gradual, broad, manifold and subtle, following the scientific revolution of the early nineteenth century. The already rapid development of Man’s power over the physical universe of the nineteenth century has been immensely accelerated by the great demands of two world wars in this century. In the days of the Renaissance there arose a new vision and a new perspective. The imaginative genius of a Copernicus and the diffusion of great works of classical antiquity gave a new zest to living and a new determination to overcome the evils which beset Man’s body and soul. This new vision achieved brilliant expression in great educational literature in which a new attempt was made

to state the importance of the individual in society, the function of political rulers and the nature of the state itself.

In the midst of the confusion and complexity of the great revolution to which I have been referring, we naturally feel that the great lights which would illumine the future have not yet arisen or that, if they have, we have not yet recognized them. One such light must arise from amongst the scientists of this generation. Whoever discharges this vital role in human affairs must provide in compelling and comprehensible form an interpretation of the nature of the physical universe which will enable eternal truths to be re-stated in a new form so that they are free from the criticism that they are intimately associated with analogies and metaphors which are already discredited, and so in tune with the largely unconscious attitude of Scientific Man to his physical environment that they are acceptable to him. The days of freemasonry in science, whether of an outwardly imposed secrecy or of an inwardly accepted incomprehensibility, are or should be over. There are signs that this is indeed realized. I have to go no further than my own University to find a professor who, speaking of the physical world, combines a clarity which comes from accuracy both of information and of method of expression with a literary grace which arises from the virtue of integrity and the good manners of desire to be comprehended. He has said:—

“. . . The nucleus of an atom and its attendant electrons are themselves very small compared with the complete atomic edifice, so that, again like the solar system, the atom is to be regarded as a rather empty structure with relatively great distances between the particles of which it is composed.

We have hitherto spoken of the structure of things in terms of atoms, nuclei, and electrons, but our knowledge of the world comes not only by touching, hearing, or smelling, which can, perhaps, be described in terms of the behaviour of atoms, but also from the sense of sight. What ‘stuff’ is responsible for the visual image of things which we receive with our eyes? To-day we speak of this stuff as radiation and we may regard it like matter itself, as being made up of discrete particles which we distinguish from atoms by the name ‘quanta’ or ‘photons.’ ”

This is patently authentic and it speaks to Man upon a matter of equal concern to all men, not as a scientist about science to other scientists, nor as a scientist condescending to those who are not scientists, but as an authority about something of significance and importance which ought to be made comprehensible to all men.

There is, therefore, much that is moving in the directions to which I have been referring. More opportunities are required to enable moves in these directions to proceed at a pace more in conformity with the changes which scientific discovery brings about. There is much that the Education Group of the Institute of Physics can do. In these opening remarks I have been chiefly trying to deal in as clear and explicit terms as possible with its opportunities.



# Some Chemical and Physical Properties of Rubber

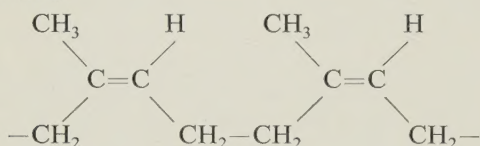
By J. MOORE, B.Sc., A.R.I.C., Research Centre, Dunlop Rubber Co., Ltd., Erdington, Birmingham

The chemical constitution and physical state of natural rubber in its viscous, high-elastic and crystalline conditions are described. It is shown that those depend on long hydrocarbon chains which are normally coiled and tangled. The high elasticity of rubber is made possible by the ability of the chains to straighten and recoil. On breaking down the cohesion between the chains by various means, rubber has some of the characteristics of a thick oil; whilst, on the other hand, certain conditions, especially those of intense cooling, cause rubber to behave in many respects like a crystalline material. This concept is then extended to a consideration of other rubber-like materials including gutta percha, some synthetic elastomers and finally the silicone rubbers.

The chemical and physical nature of rubber depends on the hydrocarbon  $(C_5H_8)_n$  which is present to the extent of 90–95% in natural rubber. By dry distillation of rubber this hydrocarbon yields, amongst other compounds, a small quantity of isoprene, a low-temperature



boiling liquid,  $\text{CH}_2=\text{C}-\text{CH}=\text{CH}_2$ . Polymerization of isoprene takes place slowly in sunlight to give first a viscous liquid and, later, a product slightly resembling rubber. This effect was first noticed by Tilden in Mason College, Birmingham, in 1900. It has now been shown that isoprene is the basic repeating unit of the rubber molecule which consists of a long chain of such units in the cis form.



The molecular weight is not precisely known and different values are obtained by different methods, but it is certainly of the order of 200,000, equivalent to approximately 3000 isoprene units.

The angle between the carbon to carbon links is approximately  $108^\circ$  and free rotation is possible at all singly-bonded carbon atoms, so that the carbon atom may take up any position on the base of a cone in relation to its adjacent one at the apex. It will readily be seen that if the carbon-carbon chain is built up according to a random distribution of all possible directions, the resulting molecule will certainly not be straight but will tend to coil back on itself in such a way that the ends are comparatively close together. Fig. 1 is an attempt to illustrate this. A length of 14 ft. of wire was bent every  $\frac{1}{2}$  in., each bend representing a carbon atom. One of six equally spaced positions on the base of the cone was selected by the throwing of a die and the wire bent accordingly. In this way a reasonably random distribution was obtained and it will be seen that the "molecule" takes on a coiled-up condition. The complexity of the actual rubber molecule will be appreciated when it is realized that this represents only a one-hundredth part of the molecule on this scale. A physical mass of rubber will therefore consist of a very large number of these coiled long-chain molecules which

are not linked together, but are considerably entangled (1). If such a mass is stressed, alteration of dimensions will result from two mechanisms:

- (a) Rotation about the singly-bonded carbon atoms, resulting in reversible chain straightening.
- (b) Slippage of one chain over another causing irreversible flow.

If this process is continued until a considerable degree of chain straightening takes place, the chains will, over

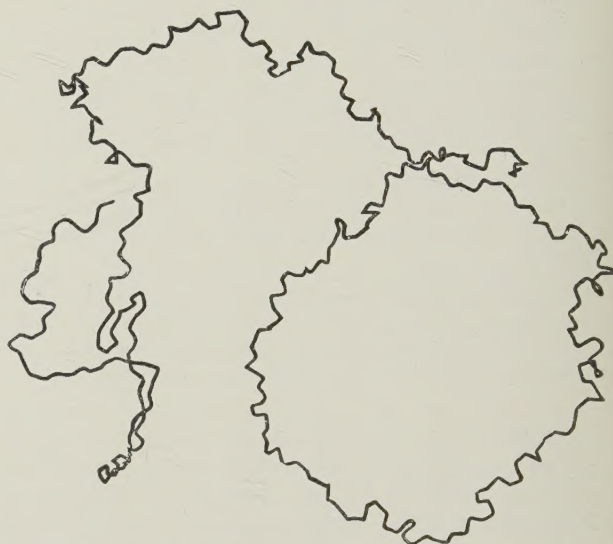


Fig. 1. Wire "model" of carbon-carbon chain molecule

small parts of their length, become parallel to each other, giving rise to a pseudo-crystallinity of a fibre-like character. This is a phase change in the system.

## RAW RUBBER

The large degree of permanent set obtained when raw rubber is stretched arises from a considerable amount of chain sliding occurring on extension. This slippage takes place despite entanglements of the chains and is considerably increased by shortening of the chains by mechanical breakage, oxidative scission, or by the introduction of plasticizers which reduce cohesion between the chains themselves. Normally, rubber is plasticized in order to aid the incorporation of necessary ingredients for rubber manufacture by milling on heavy rollers, thus causing the molecular chains to be broken down. In

this way, the rubber may become very soft and even like a thick oil. Because the deformation of raw rubber results in continuous chain sliding, the modulus and resilience under conditions of slow stressing are zero whilst the apparent viscosity coefficient is a function of applied stress.

#### RUBBER ELASTICITY

When an intimate mixture of rubber and sulphur, e.g. in the proportion of approximately 95 : 5, is heated, particularly in the presence of an accelerator and zinc oxide, vulcanization takes place. The sulphur combines with the rubber with the resultant elimination of some of the double bonds at various points in the molecule. The sulphur may either link together different parts of the same molecule or two separate molecules. The result of this is to reduce considerably the tendency of the chains to slide over each other. Although when vulcanization takes place a very small percentage of cross linkages are introduced between the chains thus bonding the two-dimensional chains into a three-dimensional network, the ability for bond rotation in the chains is substantially unaltered from the raw or unvulcanized state. Permanent set is almost eliminated and restoration of the rubber after deformation is practically complete. If more cross-linkages are introduced by using increasing percentages of sulphur, the freedom of rotation is increasingly reduced until there is reached the hard ebonite stage at which all rubber-like extensibility is lost.

The modulus of vulcanized rubber is low by comparison with crystalline materials and is affected by temperature, the interesting feature here being the fact that the modulus rises with temperature. The extensibility of rubber by bond rotation is not a consequence of an increase in the internal energy of the system, as is the case in the extension of crystalline solids, but, instead, is associated with an entropy decrease corresponding to reduction in "disorder" by the chain-straightening process. In soft vulcanized rubber at 400% extension approximately 90% of the stress arises from entropy change and only 10% from internal energy changes. The thermodynamic similarity between rubber and a gas suggests that a heat engine using rubber as the working fluid, instead of a gas, can be constructed.

Guth and James (2, 3), Wall (4, 5) and others have elaborated the thermodynamic theory of rubber elasticity and have shown good agreement between the mathematical properties of an idealized three-dimensional network of extensible molecules and the elastic behaviour of rubber at low extensions. The stress  $F$  in an extended rubber sample increases with increase in temperature according to the expression

$$F = -\frac{T\partial S}{\partial \gamma} \quad \text{or} \quad F \propto T\gamma$$

where  $F$  = tension,  $T$  = abs. temp.,  $\gamma$  = extension,  $S$  = entropy,

so that if  $F$  is constant  $\gamma$  will decrease with rise in temperature. This fact can be readily demonstrated by the rubber engine (see Fig. 2). When the rubber threads are electrically heated by the wires the modulus increases, thus lifting the weight. As the rubber approaches its maximum retraction, about 5%, the heating wires are switched off, allowing the rubber to cool. By careful balancing and the use of low-friction bearings the wheel

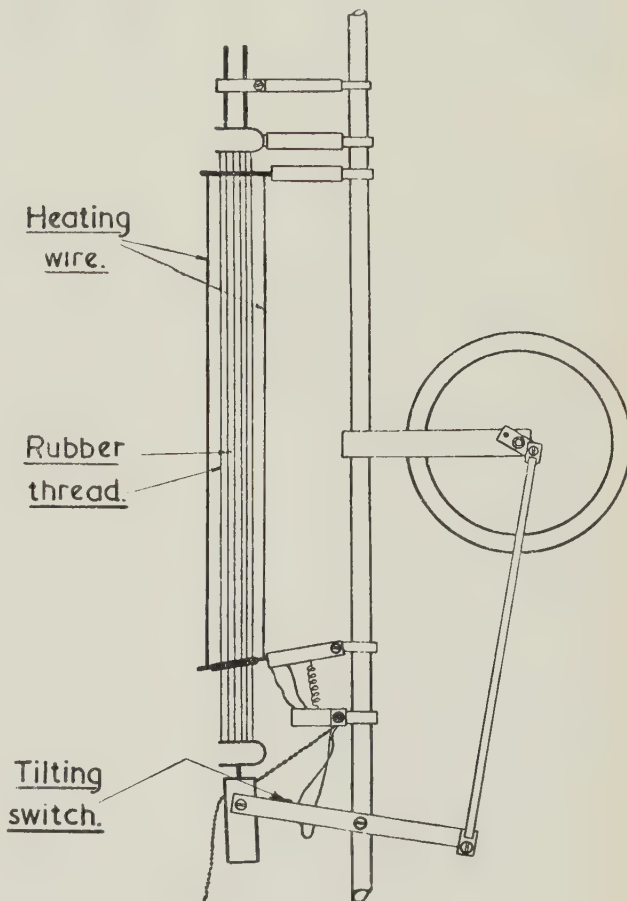


Fig. 2. Diagram of rubber engine

can be made to turn. This effect was first discovered by Joule (6), Weigand being the first to succeed in making a rubber engine work. Not only does the modulus rise with temperature but so does the resilience. This can quite readily be shown by heating a rubber ball in boiling water and comparing its rebound with a similar ball maintained at room temperature. At high temperatures the thermal motion of the molecules increases and this, as in a liquid, results in greater mobility and reduced viscosity, causing less damping of molecular movements under the forces set up by chain extension and retraction. The resilience can reach a figure of about 96–99%.

The addition of carbon blacks to rubber for reinforcement causes the resilience to fall considerably, the amount of "lowering" depending on the particle size. The smaller the particle size, the greater is the lowering



of the resilience. Even the coarser blacks cause the resilience to fall to 84% at a loading of approximately 20% (by volume) on the rubber, whilst the fine channel blacks reduce the figure to 70% and less. Thus those who are responsible for designing a rubber compound are faced with conflicting properties—(a) the gain in strength and abrasion resistance by incorporating fine carbon blacks and (b) the lowered resilience and resulting heat generation of the compound due to repeated stress cycles. Although not so marked, the variation of resilience with temperature is still present in black-loaded rubber compounds.

#### EBONITE OR VULCANITE

As previously mentioned, if all the double bonds in rubber are eliminated by combination with sulphur, ebonite is obtained. The material corresponds chemically to the empirical formula  $(C_5H_8S)_n$ . As a result of this reaction, which is exothermic, the possibility of chain straightening by bond rotation is prevented and the product is, therefore, a hard, almost metal-like, material. It is also chemically inert (due to saturation of double bonds) and is very suitable for chemically resistant vessels.

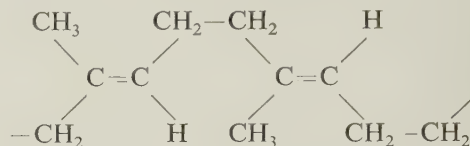
#### CRYSTALLINE STATE

X-ray work has revealed that, in ordinary rubber, small colonies of crystallinity exist. These take the form of a more or less regular pattern of molecules supposedly due to regions in which the molecular chains become parallel. This is opposite to the amorphous arrangement which one would normally associate with a material such as rubber. Under certain conditions, these colonies will grow and will represent as much as 40% of the total (7). The crystalline state is then the predominating factor in determining physical properties. This condition is brought about by cooling rubber to temperatures of  $-40^\circ\text{C}$ . when it becomes hard and brittle. The mechanical properties of the crystalline rubber are similar to those of metals—very high modulus and resilience and increased tensile strength. The high resilience of the rubber can be shown by the fact that frozen rubber, shaped in the form of a bell, will emit a musical note when struck. This state can also be brought about by "racking." Unvulcanized rubber is first calendered to a uniform sheet and then stretched. With the tension maintained, the rubber is rapidly cooled. It is then similar to a textile tape and is no longer in the high elastic stage normally associated with rubber. At normal temperatures this condition remains and samples processed as much as twenty years ago still remain racked. If, however, the temperature is raised to about  $50^\circ\text{C}$ . the rubber instantly contracts and reverts to the high elastic phase. During the process of racking the chains have been straightened by bond rotation and crystallize by cooling in that form. Even when the temperature has returned to normal the rubber remains crystalline.

#### ELASTIC PROPERTIES OF OTHER HIGH POLYMERS

The conception of elastic properties resulting from long chain molecules which are highly coiled and tangled enables the properties of other high polymers, including synthetic rubbers, to be understood.

*Gutta Percha.* This is a whitish board-like material which is obtained from the leaves of the gutta tree in a similar way to rubber. Its hardness combined with high resilience makes it a very suitable material for the outer casing of golf balls. Chemically, it consists of long chains of isoprene units linked together in the trans form,



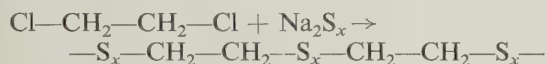
This arrangement results in a greater degree of crystallinity—a fact borne out by the properties of hardness and high resilience. On raising the temperature to about  $80^\circ\text{C}$ . the material becomes soft and rubbery. This change of state is quite analogous to the change of racked rubber to ordinary rubber. The difference lies in the range of temperature which is necessary in the two cases, gutta percha requiring a higher temperature to enable sufficient bond rotation so that the chains may straighten and recoil.

*Synthetic Elastomers.* When, for various reasons, attempts were made to make synthetic rubber, it was natural that the first material to be tried was isoprene. It was already known that this liquid would slowly polymerize to give a product resembling rubber. In spite of all attempts, however, a satisfactory product could not be made and, in fact, the rubber obtained had very poor physical properties. It was not until it was realized that a physical rather than a purely chemical imitation was needed that real progress was made. In other words, if long-chain molecules in a coiled-up state can be made, a synthetic elastomer has been achieved despite the exact chemical nature of the units of the chains. Once this concept was understood, progress was quite rapid.

The early work in this field was carried out by the Germans during the first World War for the obvious reason of national self-sufficiency. They found that by starting with dimethyl butadiene  $\text{CH}_2=\text{C}(\text{CH}_3)-\text{C}(\text{CH}_3)=\text{CH}_2$  polymerization could be brought about in solution either by the action of sodium or heat. Later it was discovered that a more satisfactory product could be obtained by co-polymerizing butadiene  $\text{CH}_2=\text{CH}-\text{CH}=\text{CH}_2$  and styrene  $\text{C}_6\text{H}_5-\text{CH}=\text{CH}_2$ . Further progress was made by carrying out the polymerization in emulsion form using peroxide types of compounds as catalysts. The reaction was carried out under pressure and gave rise to a latex from which the synthetic rubber was obtained by coagulation in much the same way as natural rubber is obtained. This was essentially the method adopted by the Americans in their

production of GR-S during the second World War. The product was manufactured in bulk and, although its physical properties were in general inferior to natural rubber, it was, nevertheless, a serviceable raw material. Recent work has produced rubbers which are a great improvement on this utilitarian material and may well become the equal of natural rubber in many important respects, particularly in abrasion resistance. Another material which illustrates the physical rather than the chemical imitation of nature is Neoprene. Here the monomer is chloroprene  $\text{CH}_2=\text{C}(\text{Cl})-\text{CH}=\text{CH}_2$ , differing from isoprene only by the replacement of a methyl group with a chlorine atom. This polymerizes under the influence of peroxides with the elimination of one of the double bonds and the resulting product is rubber-like and especially noteworthy for its oil-resistant properties and ageing resistance, particularly in the presence of light. It will be noticed that, in both these cases, one double bond remains in the polymer which can be used for the purposes of vulcanization.

Thiokol is an example of a long-chain polymer consisting of carbon and sulphur. It is prepared by the action of sodium polysulphide on ethylene dichloride

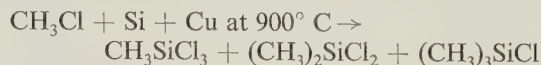


As a synthetic elastomer it has only very limited uses, since it has very poor mechanical properties. Its main property is its extreme oil resistance and, if the poor mechanical properties are not a serious disadvantage for any particular purpose, it is a very suitable material from this point of view. These products are, of course, elastic at ordinary temperatures. Many high polymers can become so under appropriate conditions. For example, polystyrene, which is a hard glass-like material at ordinary temperatures, exhibits some elastic properties at elevated temperatures—an observation reminiscent of gutta percha.

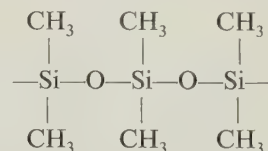
Nylon is slightly elastic at ordinary temperatures but at  $200^\circ\text{C}$ . it is quite rubbery. Its chemical structure is of the form  $\text{---NH---CO---}(\text{CH}_2)_4\text{---CO---NH---}(\text{CH}_2)_6\text{---NH---}$ . On the other hand, polyvinyl chloride  $\text{---CH}_2\text{---CHCl---CH}_2\text{---CHCl---}$  is a very hard powder when manufactured. By mixing into it a plasticizer such as dibutyl phthalate, sheets of transparent material can be obtained which again exhibit some degree of elasticity. In this case, the ability of the molecular chains to twist and straighten can only be brought about by the lubricating effect of the plasticizer. It will, however, be appreciated that the original idea of elasticity resulting from coiled molecules still holds good and it is merely a matter of achieving suitable conditions for such molecules to exhibit this property.

**Silicone Rubbers.** All the materials so far described have been purely organic and usually carbon-carbon chains with different substituent radicals attached to them. If the concept is correct, however, the possibility of other types of chain should not be ruled out.

This idea has received brilliant justification in the synthesis of the silicone rubbers which are basically built up of  $\text{---Si---O---Si---O---Si---}$  chains. Normally, silica takes on some highly crystalline state as in quartz, but long silica chains have been synthesized by the following preparation:



By purification, the second of these compounds can be treated with water to give  $(\text{CH}_3)_2\text{Si}(\text{OH})_2$  which on heating with a condensing catalyst gives:



This and similar products are long-chain polymers which are remarkably rubber-like. They can be vulcanized by special methods and yield products which remain rubbery at temperatures of  $300^\circ\text{C}$ . and higher (temperatures which would rapidly cause ordinary rubber to deteriorate and decompose). Compared with natural rubber, they are comparatively "dead," but they can be used as gaskets for high-temperature work where natural rubber would be useless. It cannot, of course, be claimed that these materials are entirely inorganic, but the basic chain of the molecule consists of silicon and oxygen and only the substituent groups contain organic material.

#### CONCLUSION

The natural rubbers and most of the synthetic rubbers consist essentially of long carbon-carbon chains with hydrogen atoms attached. Some of the synthetic rubbers may have hydrogen partly replaced by chlorine and others may possess sulphur, nitrogen or oxygen atoms in the chain itself. The carbon-carbon structure can, however, be dispensed with entirely, as in the case of silicone rubbers where the chain consists essentially of silicon atoms linked together by oxygen.

#### ACKNOWLEDGMENTS

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## ORIGINAL CONTRIBUTIONS

## The Development and Deionization Time of Heavy-Current a.c. Arcs

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[Paper received 10 March, 1949]

An account is given of some photographic observations made on long a.c. arcs at currents of 4600 to 6500 A. lasting for 0.56 to 1.12 sec. The arcs were photographed with a "still" camera and also with ciné cameras running at 20 and at 1000 pictures per sec. The development of the arc is described and it is shown that the intensely luminous cloud of vapour produced by the arc commonly persists for 0.1 to 0.2 sec. and in one instance was still visible 0.5 sec. after the current had been interrupted.

It is sometimes desirable to apply short-circuits to power systems to test the operation of protective relay equipment and tests of this nature were made on the main 132-kV grid system on 23, 24 August, 1947. The current in the arcs resulting from the short-circuits had a value of about 6000 A. and, as there is not a great deal of information available on long heavy-current a.c. arcs, the opportunity was taken to study the behaviour of the arcs with the aid of normal-speed and high-speed ciné cameras. In this way it was hoped to obtain data on the general development of the arcs and in particular on the deionization time—the time taken for the ionized gases and metallic vapours to dissipate. The deionization time is of importance in connexion with the use of high-speed reclosing switchgear. If, after a transient short-circuit, the current is interrupted, the arc extinguished and the circuit automatically switched-in again, there is very little disturbance of the supply provided that the switches are reclosed rapidly enough. Reclosing must not, however, be effected too rapidly, as the arc may restrike if the ionized vapour has not dissipated.

The discussion of high-speed reclosing is outside the scope of this paper and for further information on this subject reference should be made to electrical engineering literature such as the published work of Sporn and Müller (1) (2), Wagner (3), and Jancke (4). It is the object of this paper to describe some of the physical characteristics of long a.c. arcs.

## METHOD OF INVESTIGATION

At a suitable location on a 132-kV line route, temporary isolating and "fault-throwing" switchgear was installed to enable short-circuits to be applied to the line. The arcs were produced between two  $1\frac{1}{2} \times \frac{1}{4}$  in. mild-steel strip electrodes between which a length of 18 s.w.g. copper wire was connected to form a fuse. Single-phase, two-phase or three-phase short-circuits of various durations could be applied. It should be emphasized that the observations of the arcs were

subsidiary to the primary object of the investigations and the various test conditions were arranged with the object of elucidating the operation of the protective relays under certain practical conditions. The arc currents were measured with high-speed ammeters supplied through current transformers. These ammeters are rectifier-type instruments with high torque and heavy damping so that the movement attains full deflexion in 0.1 sec. A "slave" pointer is fitted to indicate the maximum deflexion. The duration of the currents was obtained from oscillograph records and also from the ciné films.

*Photographic Technique.* Photographs of the arcs were taken with a 16 mm. high-speed ciné camera, a 16 mm. normal-speed ciné camera and an ordinary twin-lens  $2\frac{1}{4}$  in. square reflex camera. The high-speed ciné camera (5) (6) was used to photograph the arc broadside-on at a mean picture speed of 1000 frames per sec., that is, at 10 frames per half-cycle of the 50 c/s current. The high-speed camera was used with the object of photographing the most intense regions of the discharge and accordingly a short exposure time was employed—1/5000 sec. at f. 16 using Super XX film. The film speed is not constant, but timing marks are photographed on the film every one msec. and enable time intervals to be accurately measured. The normal-speed ciné camera was set up in a direction at right angles to that of the high-speed camera so that an end-on view of the arcs was obtained. The picture frequency was 20 frames per sec. corresponding to an effective exposure of 1/40 sec. Super X film was used with a lens aperture of f. 16 and an  $\times 2$  yellow filter. The still pictures were taken on Panatomic X film with an exposure of 1/100 sec. at f. 22. This camera was mounted beside the normal-speed ciné camera.

## RESULTS OF INVESTIGATIONS

Six short-circuits were applied with arc currents of from 4600 to 6500 A. and durations of from 0.56 to 1.12 sec.; other data, including the deionization times



obtained from the high-speed film, are given in the Table.

*Test conditions and deionization times*

Test No.	Type of short-circuit	Arc gap in cm.	Current in A.	Current duration in sec.	Deionization time in sec.	
					(a)	(b)
1	3-phase	71	6500	1.12	0.1	0.16
2	1-phase	71	6000	0.74	0.1	0.18
3	1-phase	200	5750	0.56	0.13	0.31
4	2-phase	182	5500	0.83	0.3	0.40
5	1-phase	182	5500	0.82	0.1	0.23
6	3-phase	182	4600	0.80	0.17	0.33

The arc voltage was not measured directly, but voltage measurements were made at a transforming station



Fig. 1. Three-phase short-circuit (Test 1)—photograph taken with still camera

4 miles from the test site and it is estimated from these measurements that the total voltage drop in the arc was only 3 or 4 kV (r.m.s.). This corresponds to an arc voltage of the order of 10 or 20 V/cm. depending on the arc length for which a rough estimate only could be made. The arc resistance was about  $0.7 \Omega$ .

Fig. 1, a photograph of a three-phase short-circuit taken with the still camera, gives a general view of the first test—the high-speed ciné camera was located to the right of the photograph. The photographs were all taken in the bright daylight of a fine August afternoon and the extreme luminosity of the arcs is very apparent.

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In Fig. 1, for example, the daylight illumination of the test area is small compared to that due to the light from the arcs. In the ciné films the sky appears as a dark background to the arcs and, in one of the still-camera pictures the sun, slightly veiled by haze, was included in the field of view and appeared to be of similar brightness to the arc. This indicates an arc temperature of the order of  $6000^\circ \text{K}$ .

*Development of the Arc.* The development of the arc can best be described with the help of a sequence of selected stills from the high-speed film. Fig. 2 shows the growth and decay of the arc in Test 5. In the first

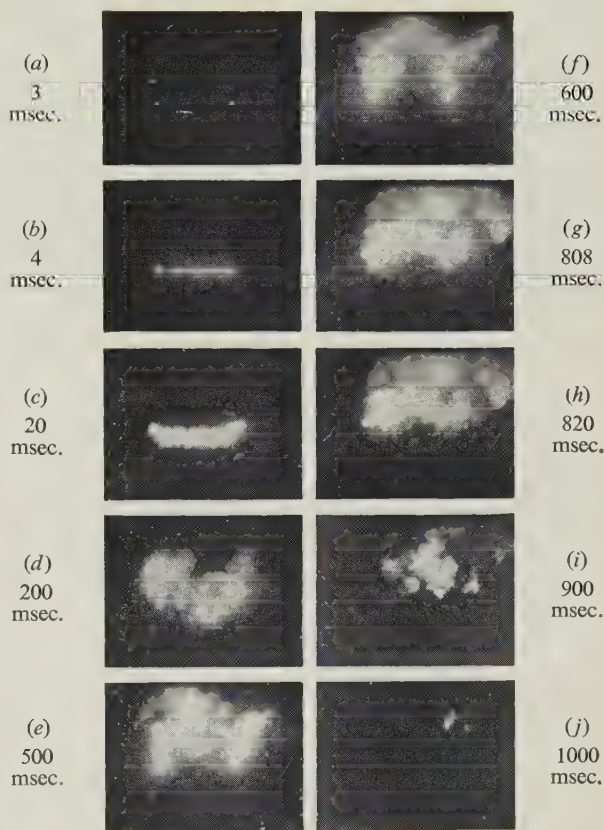


Fig. 2. Growth and decay of arc—sequence of stills from high-speed ciné film (Test 5)

few msec. discharges appear at the two points where the fuse wire is connected to the electrodes and Fig. 2(a) shows the appearance of these discharges 3 msec. after they were first observed. One msec. later the wire fuses and presents a beaded appearance as in Fig. 2(b). This phenomenon has been described by Baxter (7) and by Gantenbein (8). The wire is suddenly heated to its melting point and the resulting long fluid cylinder is unstable and breaks up into drops. Arcs then form between the molten globules giving rise to the beaded effect shown in the figure. This condition lasts for a fraction of one msec. and the multiple arcs then give

place to a single discharge along a straight line between the electrodes. It may be remarked that the calculated time to melt an 18 s.w.g. copper wire with a current of 5500 A. (r.m.s.) is from  $\frac{1}{2}$  to 4 msec. depending on the part of the cycle at which the circuit is closed. After 20 msec. the arc is fully established as in Fig. 2(c). This photograph was taken at a time when the current was passing through zero and it is noteworthy that, even in

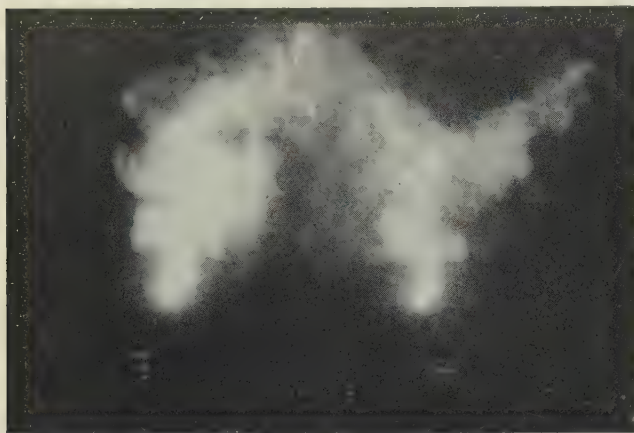


Fig. 3. Enlargement of Fig. 2(f) showing arc channels within vapour cloud

its early stages, the arc never appears to be extinguished at zero current.

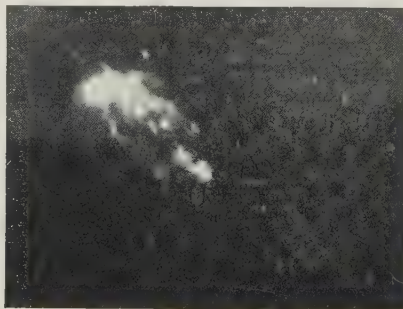
Throughout most of the film the alternations of the current are marked only by a pulsation in the general luminosity of the discharge and the actual discharge path is obscured by the relatively permanent cloud of glowing gas and vapour as in Figs. 2(d) and 2(e). On some parts of the film, however, glimpses of the actual arc channel can be seen in the form of sinuous discharges threading their way through the cloud of vapour. Owing to their motion, these discharges are most noticeable when the film is projected, but they can be seen in Fig. 2(f) and more clearly in the enlargement (Fig. 3). The arc channel appears to have a diameter of a few cm. and its length is of the order of 3 m. The cloud of ionized gas and vapour increases steadily in size as the arc develops

and Fig. 2(g) shows its appearance just before the current is interrupted. After current interruption, recombination takes place and the cloud of vapour dissipates gradually—Figs. 2(h), 2(i) and 2(j). The final stages of the same arc (Test 5) as recorded by the normal-speed ciné camera are shown in Fig. 4. In this case the film was more fully exposed and the camera had a shorter-focus lens so that the background details and the whole extent of the arc can be seen. The arc rose to a height of about 4 m. and the light wind caused it to drift sideways as shown in Fig. 4(a), to such an extent that it enveloped the downcoming connexions to one of the outer phases and, as the oscillograph record showed, developed into a two-phase short-circuit 0.1 sec. before the current was interrupted. Figs. 4(b) and 4(c) show the deionization of the cloud of vapour, the luminosity being more persistent in the higher regions of the cloud.

*Deionization Time.* As might be expected, the deionization time was found to be a rather indefinite concept and, accordingly, two values have been given in the Table: (a) the time after current interruption for which incandescent vapours persist in such quantities that an arc would be likely to restrike and (b) the total time from current interruption until the last traces of incandescent vapour just disappear. The deionization times obtained from the normal-speed ciné film were in general agreement with the figures given in the Table although they tended to be rather greater, possibly owing to the fact that the normal-speed film was exposed much more fully than the high-speed film. Although the deionization times vary considerably for apparently similar arcs, it is clear that the longer times are associated with the greater gap lengths. By taking the mean of the values in the Table and taking into account the results from the normal-speed film, it is concluded that with a current of about 6000 A. large clouds of incandescent vapour persist for 0.1 sec. for a 71 cm. gap and for 0.2 sec. for a gap length of 182 to 200 cm. The corresponding times for the complete dissipation of the incandescent vapour are 0.2 and 0.4 sec. respectively. The maximum deionization time observed (0.4 sec. on the high-speed and 0.5 sec. on the normal-speed film) occurred in Test 4, and Fig. 5, a still from the normal-speed film, shows the large clouds of incandescent vapour



(a) 0.83 sec.



(b) 0.9 sec.



(c) 1.0 sec.

Fig. 4. Deionization of arc—sequence of stills from normal-speed ciné film (Test 5)





Fig. 5. Clouds of incandescent vapour 0.2 sec. after current interruption—still from normal-speed ciné film (Test 4)

0.2 sec. after current interruption. The maximum height of the cloud of vapour is about  $4\frac{1}{2}$  m.

## ACKNOWLEDGMENT

The author is indebted to Mr. E. R. Davies and Mr. G. A. Jones of the Research Laboratory of Kodak Ltd. for valuable assistance in connexion with the high-speed ciné photographs.

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## The Measurement of Opacity and Reflectivity for Printing Papers

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A form of reflectometer for measuring the "show through" tendencies of printing papers is described. Its grading of a series of nineteen papers covering a wide range of opacity is in good agreement with the judgment of 173 trade observers. The instrument can also be used for measuring reflexion factors. "Diffuse illumination, normal viewing" and "normal illumination, diffuse viewing" can both be used and are shown experimentally to be practically equivalent, but the latter is more convenient. The opacity figures can be converted to the U.S. "TAPPI Opacities" by means of an equation due to Judd; the results obtained are in good agreement with measurements made on the American Bausch and Lomb Opacimeter.

As a printing material, paper suffers from the drawback that it is translucent. This may cause printed matter to appear through the reverse side of the sheet and the resultant "show through" is objectionable, particularly when both sides of a sheet are printed. Possible methods of measuring the opacity of paper have been discussed at some length in a previous paper (1). A measure of "show through" tendencies is required and this is not the same as the transmission density in the photographic sense. The instrument to be described has been designed in collaboration with Hilger and Watts Ltd. (Hilger Division) and adheres to the recommendations put forward in the paper mentioned.

### METHOD OF MEASUREMENT

The opacity of the paper measured is expressed in terms of contrast ratio or printing opacity. Contrast ratio is defined as the percentage ratio of the reflexion factor ("brightness") of the sample, when backed with a black surface, to its reflexion factor when backed with a standard white surface under the same conditions of

illumination and viewing. The percentage is zero for a perfectly transparent paper and 100% for one which is quite opaque. All real papers give values lying between these extremes. The chief drawback of the contrast-ratio method lies in the difficulty of preparing and maintaining a satisfactory standard white surface: this difficulty is avoided in the printing opacity method. Printing opacity is measured in exactly the same way as contrast-ratio except that the standard white backing is replaced by a thick wad of the paper under examination. A "thick wad" is defined as one of such thickness that its doubling makes no measurable difference to the reflexion factor of the outer surface: with normal printing papers a wad of ten to twenty sheets is sufficient. For all ordinary opacity determinations, the use of printing opacities is preferred. If, however, very small samples are sent for test it may be impossible to make a sufficiently thick wad for the backing, in which case contrast-ratios should be used, a convenient backing being a block of magnesium carbonate of reflexion factor about 97%. Printing opacity figures are higher than the corresponding

contrast-ratios, but are inter-related, as will be shown later.

It will be inferred from the foregoing that the opacimeter is essentially a photometer capable of comparing the reflexion factors of the black-backed and self-backed (or the white-backed) specimens under carefully specified conditions of illuminating and viewing. The colour of the illuminant, the spectral sensitivity of the photometer and the geometry of the illuminating and viewing systems are all important and the following conditions have been adopted as standard:

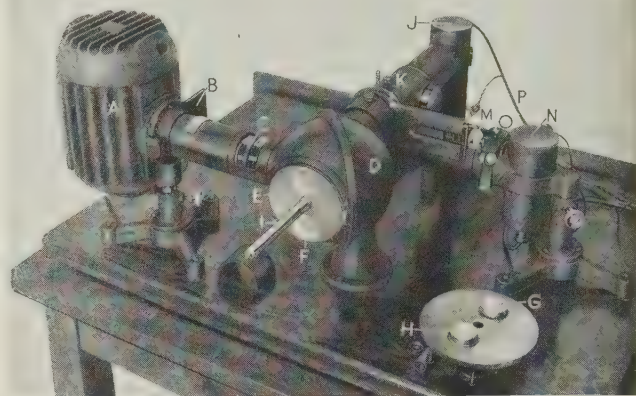
- (1) Illuminant: Source B as specified by the 1931 Commission Internationale de l'Éclairage.
- (2) Photometer: Spectral sensitivity the same as the photopic visibility curve of the C.I.E. Standard Observer.
- (3) Geometry: Specimen "viewed" normally and illuminated by completely diffused light. The alternative method of illuminating the specimen normally and "viewing" it diffusely by means of an integrating sphere offers some advantages from the point of view of instrumentation and the two systems are generally supposed to be equivalent. The instrument now described has been designed for experimental purposes and allows either method of illuminating and viewing to be used at will; but this convertibility is not intended to be a feature of the opacimeter in its final commercial form.

It follows that the instrument will measure not only printing opacities but also the reflexion factors of the black- and self-backed samples under the stated conditions of illuminating and viewing. In this way it will give an estimate of the lightness ("brightness") of papers as seen by a normal observer in diffused daylight, though it must be stated that this estimate will not always agree with a papermaker's subjective estimate of the "brightness" of a paper, which involves colour as well as reflectivity.

#### CONSTRUCTION OF INSTRUMENT

The instrument follows normal photometric principles and its construction will be apparent from the figure. Light from a tungsten-filament lamp operated at a colour temperature of 2848° K. enclosed in a housing *A* impinges on the wall of a 6 in. integrating sphere *D* via a shutter *B* and the two C.I.E. liquid filters *C*. In front of the integrating sphere are two apertures *E* and *F* into which fit the specimen holders *G* and *H*. The positions of *G* and *H* may be reversed by rotating the disk carrying them about an axis *I*. The specimens themselves are kept gently pressed against a flange by a spring and plunger arrangement so that a circle 1 in. in diameter is exposed. A convex lens throws an image of the specimen in aperture *E* on to the cathode of a vacuum photocell *J* provided with a deep yellow correction filter inserted at *K* and the amount of light reaching the cell is controlled by a rectangular diaphragm *L* of 25 mm. maxi-

mum aperture mounted immediately in front of this convex lens and governed by a micrometer screw worked by a handle and indicating drum *M*. The specimen in aperture *F* is viewed by a comparison photocell *N* with yellow correction filter *O* and the amount of light reaching this cell is controlled by an iris diaphragm at *P* actuated



Opacity and reflectivity meter for printing papers

by a small lever (not visible in the figure). Leads from the two cells *J* and *N* are taken to a mains-operated d.c. amplifier. The equipment is not particularly sensitive to changes in mains voltage but a constant voltage transformer is included. The arrangement depicted conforms to the "diffuse illumination, normal viewing" method. In use, a single sheet of the paper under test, backed with a black disk, is introduced at *E* and a white porcelain disk at *F*. The light from the lamp is cut off by closing shutter *B*, the diaphragm *L* is opened to its full extent and the cells are balanced in the dark by adjustment of the amplifier controls. Balance is shown when there is no deflexion of a centre-zero microammeter. Shutter *B* is then opened and balance restored by adjustment of diaphragm *P*. Thirdly, the black-backed sheet at *E* is replaced by a thick wad (for printing opacities) or a white-backed sheet (for contrast ratios) and the balance again restored by closing diaphragm *L*. The ratio of the aperture of *L* (shown by *M*) to its full aperture gives the measure sought; in practice, since full aperture is 25 mm., one simply multiplies the reading of *M* by 4. In the alternative arrangement ("normal illumination, diffuse viewing"), the positions of light *A* and cell *J* are interchanged. Cell *N* no longer views a white disk introduced at aperture *F* but is instead arranged to catch some of the light of the lamp by means of a reflecting plate mounted in the tube near *K*. The response of cell *N* is thus proportional to the total incident flux whilst the response of *J* is proportional to the total reflected flux collected by the integrating sphere *D*. The optical system is such that an image of the lamp filament is thrown on to the sample at *E*. In use, a single black-backed sheet is introduced at *E* and a thick wad (or white-backed sheet) at *F*, and the instrument is, as



previously, balanced with  $L$  at full aperture. The positions of the black-backed and self-backed sheets are then reversed by rotating the mounting disk about the axis  $I$ , balance being restored by closing the diaphragm  $L$ . The opacity is calculated as before.

It is evident that the reflectivity of a paper may be readily determined by mounting a thick wad of the paper and a standard magnesium oxide surface in positions  $E$  and  $F$  respectively and proceeding as before. A null method is thus used and the accuracy of measurement depends largely on the uniformity of the light flux passing through the rectangular diaphragm  $L$ . The makers have given particular attention to this point and we have checked it by masking out rectangular portions of this aperture and observing the effect on the results after rebalancing the system. When the printing opacity of a paper was determined with the aperture progressively reduced from one side to  $\frac{4}{5}$ ,  $\frac{3}{5}$ ,  $\frac{2}{5}$  and  $\frac{1}{5}$  of its full extent, the values found agreed to within 0.1% after correcting for a small zero error in the micrometer scale. We are therefore satisfied that no important error arises from this source. Repeat observations on a single specimen of printing opacity 83% showed a standard deviation of 0.16 "one-per-cent-units" from the mean. In a group of 10 observations, therefore, errors of setting will not introduce an error in the mean of more than 0.10 unit in 19 cases out of 20. Since differences of 0.2 in printing opacity are commercially insignificant, this degree of precision in the measurements is adequate.

#### COMPARISON OF ALTERNATIVE METHODS OF ILLUMINATING AND VIEWING

We were able to secure for the experimental work described herein a set of 19 papers ranging from almost complete opacity down to 72% printing opacity (almost a tracing paper); 110 sheets of each were available, of which every eleventh sheet was retained for measurement, whilst the remaining hundred were circulated for the visual grading test described later. From each of the ten sheets thus sampled, two circles of size suitable for the opacimeter were cut with centres along one of the diagonals, giving twenty specimens in all, which were then measured with the instrument in the "diffuse illumination, normal viewing" position. A further twenty samples were next cut with centres along the remaining diagonals of the sheets and measured in the "normal illumination, diffuse viewing" position—fresh specimens being employed in order to avoid effects of possible contamination of the surfaces during the previous measurement. The results of all these measurements are summarized in Table 1, which gives the mean printing opacities of the 19 papers of the series according to the alternative methods of illuminating and viewing and the standard deviations ( $\sigma$ ) of the twenty measurements in each set. An examination of this Table shows there is nothing systematic in the differences in printing opacity given by the two methods, sometimes the one giving the higher value, sometimes the other. The

significance of these differences may readily be established on the assumption that a difference is not significant unless it exceeds  $3\epsilon_{12}$  where  $\epsilon_{12}^2 = (\sigma_1^2 + \sigma_2^2)/20$ . On this basis, only papers Nos. 6, 9 and 13 show significant differences, while Nos. 3 and 5 may be classed as doubtful, the differences being about  $2\frac{3}{4}\epsilon_{12}$ . These differences may be due both to sampling variations in the papers and errors in setting the instrument, as well as to the possible effects of altering the conditions of illuminating and viewing. Even with the differences classed as significant, however, there is nothing to suggest that the alteration of conditions produces any systematic change in the readings as the results for papers Nos. 9 and 13 show. According to Farebrother (2), differences in contrast ratio of less than 0.6 cannot be resolved by eye and differences of 0.3 through sampling variations are certainly possible. The only serious difference observed is for paper No. 6. This, however, is the most transparent of the series and such papers frequently show wide variations in opacity from one part of a sheet to another so that a sampling variation of 1.1 is not impossible. From the evidence furnished by Table 1 we conclude that the alternative methods of illuminating and viewing are sensibly equivalent as far as the grading of the papers is concerned and, accordingly, it matters little which is used. In practice it is slightly more convenient to use the "normal illumination, diffuse viewing" arrangement, which will therefore be adopted as standard in the final form of the instrument. This will enable certain simplifications to be made in the design.

Table 1. Mean printing opacities, according to alternative methods of illuminating and viewing, and standard deviations

Paper No.	Diffuse illumination, normal viewing		Normal illumination, diffuse viewing		Significance
	Printing opacity	$\sigma_1$	Printing opacity	$\sigma_2$	
1	97.1	0.40	97.0	0.43	No
2	98.0	0.13	98.0	0.46	No
3	95.6	0.19	95.9	0.41	Doubtful
4	83.7	0.65	83.9	0.88	No
5	99.2	0.19	99.5	0.46	Doubtful
6	73.0	0.70	71.9	0.87	Yes
7	93.5	0.39	93.6	0.38	No
8	95.6	0.29	95.6	0.56	No
9	100.0	—	99.7	0.22	Yes
10	88.5	0.51	88.4	0.79	No
11	93.6	0.29	93.5	0.76	No
12	95.3	0.24	95.4	0.50	No
13	98.9	0.22	99.4	0.35	Yes
14	94.9	0.57	95.3	0.74	No
15	96.1	0.22	96.3	0.55	No
16	75.9	0.62	75.6	1.24	No
17	92.8	0.39	92.9	0.62	No
18	84.6	0.60	84.5	1.08	No
19	98.2	0.14	98.4	0.44	No

#### COMPARISON WITH VISUAL GRADING

We consider it important that the opacimeter should grade papers in the same order as the average trained observer and therefore circulated 100 sets of the nineteen

papers among printers and paper mills, with the request that the papers should be graded by eye in order of opacity. Up to the time of closing the lists, 173 independent estimates had been received. The distribution

similar papers but should not be given any quantitative significance, since the observers were asked only to rank the papers and not to estimate the magnitude of the differences between them. The visual ranking of the

Table 2. *Grading of paper by visual viewing*

Paper No.	Rank																		
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
9	160	10	1	1	—	—	—	—	—	—	—	—	—	—	1	—	—	—	—
13	18	87	64	2	1	1	—	—	—	—	—	—	—	—	—	—	—	—	—
5	8	58	83	22	1	—	—	1	—	—	—	—	—	—	—	—	—	—	—
19	4	17	13	116	20	2	—	1	—	—	—	—	—	—	—	—	—	—	—
2	2	2	2	21	121	8	8	1	3	1	—	1	—	2	—	—	1	—	—
1	—	1	1	—	12	106	21	15	7	3	2	2	2	—	1	—	—	—	—
15	—	—	—	1	3	16	74	33	24	7	11	1	2	1	—	—	—	—	—
3	—	—	2	—	5	11	38	36	32	35	7	6	—	1	—	—	—	—	—
8	—	—	—	—	2	9	23	39	25	46	14	6	6	2	1	—	—	—	—
12	—	—	—	—	7	7	19	26	57	43	10	1	2	1	—	—	—	—	—
14	—	—	—	1	2	8	6	5	11	27	66	20	15	8	1	3	—	—	—
11	—	—	—	—	—	—	2	2	3	2	15	48	51	45	3	2	—	—	—
7	—	—	—	—	—	—	—	1	4	4	19	41	51	36	10	4	3	—	—
17	—	—	—	—	—	—	—	1	2	7	19	44	36	54	7	1	2	—	—
10	—	—	—	—	1	—	2	—	—	3	1	4	8	11	123	14	5	1	—
18	—	—	—	—	—	—	—	—	—	2	—	2	—	1	13	102	52	1	—
4	—	—	—	—	—	—	1	—	—	—	1	1	1	6	11	50	102	—	—
16	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	1	142	30
6	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	1	34	138

of the "voting" is summarized in Table 2. The gaps between the papers in the series were intentionally not uniform and some were narrow so that visual distinction between them was difficult: this is reflected in the "voting." The papers are ranked from the most opaque,

papers is given in Column 3 of Table 3. Column 4 gives the printing opacities as measured in the opacimeter (mean of both methods of illuminating and viewing) and column 5 the ranking deduced therefrom. Satisfactory agreement between visual and printing opacity

Table 3. *Visual and opacimeter gradings*

Paper No.	Visual grading		Printing opacity		Reflectivity against magnesium oxide	Contrast ratio		Contrast ratio on Bausch and Lomb opacimeter	Pausch and Lomb ranking
	Mean "vote"	Final ranking	Mean value	Ranking		89% backing	87% backing		
9	1.16	1	99.8 <sub>5</sub>	1	63.8	99.1	99.2	99.3	1
13	2.34	2	99.1 <sub>5</sub>	3	80.0	98.5	98.7	98.4	2
5	2.73	3	99.3 <sub>5</sub>	2	74.2	98.5	98.7	98.3	3
19	3.82	4	98.3	4	81.3	97.3	97.6	97.8	4
2	5.24	5	98.0	5	63.0	94.3	94.8	96.0	5
1	6.64	6	97.0 <sub>5</sub>	6	74.5	94.5	95.0	94.5	6
15	7.84	7	96.2	7	72.4	92.6	93.2	93.2	7
3	8.39	8	95.7 <sub>5</sub>	8	73.6	92.2	92.8	92.5	8
12	8.80	9	95.3 <sub>5</sub>	10	73.7	91.7	92.3	92.2	9
8	9.04	10	95.6	9	70.4	91.3	91.8	91.8	10
14	10.68	11	95.1	11	70.2	90.4	91.1	90.5	11
11	12.63	12	93.5 <sub>5</sub>	12	71.2	88.3	89.1	89.3	12
17	12.79	13	92.8 <sub>5</sub>	14	75.5	88.6	89.4	88.9	13
7	12.82	14	93.5 <sub>5</sub>	12	71.3	88.3	89.1	88.8	14
10	14.68	15	88.4 <sub>5</sub>	15	74.2	82.6	83.6	84.2	15
18	16.10	16	84.5 <sub>5</sub>	16	75.5	78.5	79.5	78.8	16
4	16.33	17	83.8	17	74.4	77.3	78.2	78.1	17
16	18.17	18	75.7 <sub>5</sub>	18	75.0	68.7	69.8	70.5	18
6	18.80	19	72.4 <sub>5</sub>	19	77.0	66.3	67.4	67.4	19

No. 1, to the least opaque, No. 19. The order can be decided at a glance in many cases. Column 2 of Table 3 contains the means of the voting distributions of Table 2 and these means are useful in deciding the ranking of

gradings is shown, the rank correlation coefficient being 0.9921. The reversal of papers Nos. 13 and 5, 12 and 8 is unimportant, as the first pair are of very high opacity and the second pair are barely distinguishable by eye.



The most serious discrepancy between visual and instrumental gradings exists between papers Nos. 19 and 2 for, although the printing-opacity figures grade the papers in the right order, these figures are close together whereas the papers can be easily distinguished. We shall return to this later.

#### CONVERSION OF PRINTING OPACITY VALUES TO TAPPI OPACITY

The method of measuring opacity adopted in the new opacimeter is similar in several respects to the Standard Method adopted by the U.S. Technical Association of the Pulp and Paper Industry (TAPPI) whose present Standard (T425m-44—corrected August 1944) may be summarized as follows:

- (1) Opacities to be expressed as contrast ratios
- (2) Diffuse illumination, normal viewing or normal illumination, diffuse viewing
- (3) Black backing less than 0.5% reflexion factor
- (4) White backing 91.5% reflexion factor against magnesium oxide (i.e. 89% absolute reflexion factor); magnesium carbonate covered by a glass cover-slip is suitable
- (5) Colour temperature of illuminant 2400°–2800° K.
- (6) Human eye or its equivalent

Our instrument operates at a higher colour temperature, 4800° K., and printing opacities are used instead of contrast ratios but the methods are otherwise the same. Allowance cannot readily be made for the difference in colour temperature, though in general our conditions will give the higher values. Conversion of printing opacities to contrast ratios can be made, however, by means of Judd's equation (3)

$$C_r = \frac{C}{1 - (1 - C)[1 - R(1 - C)/(1 - RC)]}$$

where  $C$  is the contrast ratio obtained with an ideal, perfectly-reflecting white backing and  $C_r$  is the contrast ratio under similar conditions with a backing of reflexion factor  $R$ . The printing opacity is the contrast ratio with a backing having the reflectivity of the sample and hence, if printing opacity and reflectivity are measured,  $C$  can be calculated. From this the contrast ratio for a backing of 89% reflexion factor can be determined. This has been done in Column 7 of Table 3, using the measured values of reflectivity against magnesium oxide given in Column 6.

Column 9 of Table 3 reproduces figures for the same papers obtained on a Bausch and Lomb Opacimeter. This instrument, not generally available in this country, conforms approximately to TAPPI specification. Comparison of columns 7 and 9 shows that the agreement between calculated and measured values of the TAPPI opacity is satisfactory. Still better agreement

may be obtained if the contrast ratio is computed for a backing of reflexion factor 87% (the reflexion factor of the backing used in the opacimeter employed here has not been determined and may be on the low side. These values are given in column 8 and comparison of columns 8 and 9 will show that in most cases the agreement is very close. Such differences as remain may reasonably be attributed to sampling and the different colour temperature used. Column 10 gives the ranking of the papers according to the Bausch and Lomb opacimeter. Agreement with the visual ranking is complete, the rank correlation coefficient being +1.0000. Though both printing opacity and TAPPI opacity give very satisfactory grading of the papers, the Bausch and Lomb figures appear to be slightly preferable. In particular, the grading of papers Nos. 19 and 2 is more satisfactory by TAPPI opacity. It must be remembered, however, that in most instances the observers graded their papers by placing single sheets over a printed test card or over a page of a book. These are contrast ratio conditions. Shortage of material precluded the formation of thick wads. On the other hand, when paper is bound up in a book or newspaper it is self-backed and the opacity is judged under printing opacity conditions. Although paper No. 2 is distinctly more translucent than No. 19, as shown by contrast ratio, yet in practice its "show-through" tendencies are not markedly greater than No. 19 on account of its much lower reflectivity. It is difficult to decide the smallest difference in printing opacity or contrast ratio perceptible under favourable conditions of observation. Examination of Table 3, however, suggests that this is not less than 0.3 unit and in ordinary commercial work this may well rise to the 0.6 unit suggested by Farebrother (2).

#### ACKNOWLEDGMENTS

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# The Measurement of the Effectiveness of Social Policies in the United Kingdom

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*Abstract*—This paper discusses the measurement of the effectiveness of social policies in the United Kingdom.

The paper discusses the measurement of the effectiveness of social policies in the United Kingdom. It begins by defining the concept of effectiveness and then discusses the various methods used to measure it. The paper then discusses the results of the measurement and the implications for policy-making.

The measurement of the effectiveness of social policies is a complex task. It requires the use of a variety of methods, including surveys, experiments, and quasi-experiments. The results of the measurement can be used to inform policy-making and to evaluate the impact of social policies on different groups of people. The paper discusses the various methods used to measure the effectiveness of social policies and the results of the measurement. It also discusses the implications for policy-making and the need for further research in this area.

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The solution of equation (1) satisfying the boundary conditions (3) and (4) is

$$\phi(x) = (A/2)\{e^{i\omega x[\rho/(E+i\omega\eta)]^{\frac{1}{2}}} + e^{-i\omega x[\rho/(E+i\omega\eta)]^{\frac{1}{2}}}\} \quad (5)$$

$$\text{Since } i\omega x[\rho/(E+i\omega\eta)]^{\frac{1}{2}} = (x\omega/X)(\rho/2)^{\frac{1}{2}}[(X-E)^{\frac{1}{2}} - i(X+E)^{\frac{1}{2}}] \quad (6)$$

$$\text{where } X^2 = E^2 + \omega^2\eta^2$$

we have

$$\xi = A\left[\{\cos(x\omega/X)[2\rho(X-E)]^{\frac{1}{2}} + \cos(x\omega/X)[2\rho(X+E)]^{\frac{1}{2}}\}/2\right]^{\frac{1}{2}} \cos(\omega t + \delta) \quad (7)$$

where  $\tan \delta = \tan a \tan b$ ,  $a$  and  $b$  being the real and imaginary parts of equation (6).

The phase  $\delta$  is dependent on  $x$  and is zero when  $x = 0$ .

If the force at the driving end  $x = l$  is expressible as  $F \cos(\omega t + \psi)$ ,  $F$  being the maximum stress, the equation of motion at  $x = l$  is

$$E\partial\xi/\partial x + \eta\partial^2\xi/\partial x\partial t = F \cos(\omega t + \psi) \quad (8)$$

and substituting for  $\partial\xi/\partial x$  and  $\partial^2\xi/\partial x\partial t$  in equation (8) gives

$$\begin{aligned} F \cos(\omega t + \psi) &= (\omega A)\left[\rho X\{\cosh^2(l\omega/X)[\rho(X-E)/2]^{\frac{1}{2}}\right. \\ &\quad \left.- \cos^2(l\omega/X)[\rho(X+E)/2]^{\frac{1}{2}}\}\right]^{\frac{1}{2}} \cos(\omega t + \delta_1 + \phi) \end{aligned}$$

where  $\psi = \delta + \phi$ ,  $\tan \delta_1 = \coth a \tan b$  at  $x = l$ , and  $\tan \phi = [(X+E)/(X-E)]^{\frac{1}{2}}$ . The amplitude of vibration at the free end  $x = 0$  is therefore given by

$$\begin{aligned} A &= (F/\omega)\left[\rho X\{\cosh^2(l\omega/X)[\rho(X-E)/2]^{\frac{1}{2}}\right. \\ &\quad \left.- \cos^2(l\omega/X)[\rho(X+E)/2]^{\frac{1}{2}}\}\right]^{-\frac{1}{2}} \quad (9) \end{aligned}$$

This expression is valid for all values of  $X$  and therefore  $\eta$ .

If  $\eta\omega < E$ , terms containing  $X$  can be expanded and equation (9) becomes

$$\begin{aligned} A &= [F/\omega(\rho/E)^{\frac{1}{2}}]\left[\cosh^2(l\omega^2\eta/2E)(\rho/E)^{\frac{1}{2}}\right. \\ &\quad \left.- \cos^2 l\omega(\rho/E)^{\frac{1}{2}}\right]^{-\frac{1}{2}} \quad (10) \end{aligned}$$

If  $\eta^2\omega^2 < 0.008E^2$ , terms in  $\omega^2\eta^2/E^2$  can be neglected compared to unity to an accuracy of 1%. This approximation has been made in equation (10). Inspection of equation (10) shows that  $A$  will have maximum values when  $\cos^2 l\omega(\rho/E)^{\frac{1}{2}} \simeq 1$

$$\text{or } l\omega(\rho/E)^{\frac{1}{2}} \simeq n\pi \quad (11)$$

$$n = 1, 2, 3$$

Expanding  $\cos^2 l\omega(\rho/E)^{\frac{1}{2}}$  in terms of  $[n\pi - l\omega(\rho/E)^{\frac{1}{2}}]$ , and  $\cosh^2(l\omega^2\eta/2E)(\rho/E)^{\frac{1}{2}}$  it can be shown that maximum values of  $A$  occur when

$$\omega = (n\pi/l)(E/\rho)^{\frac{1}{2}}(1 - \delta) \quad (12)$$

where  $\delta = 3\omega^2\eta^2/4E^2 \leq 0.006$ .

Equation (11) is therefore accurate to within 1% and can be used to determine  $E$ .

The maximum amplitude  $A_{max}$  is given by substituting the value of  $\omega$  given by equation (11) into equation (10). If the resonant frequency is altered by an amount  $\Delta/2\pi$  so that the amplitude becomes  $A_{max}/\sqrt{2}$ , substitution in equation (10) leads to the following relation.

$$2(l^2\omega_n^4\eta^2\rho/4E^3) = (1 + \Delta/\omega_n)^2 [l^2\eta^2\rho\omega_n^4(1 + \Delta/\omega_n)^4/4E^3 + \Delta^2 l^2\rho/E] \quad (13)$$

where  $\omega_n$  refers to the resonant frequency and  $\cos^2 l(\rho/E)^{\frac{1}{2}}(\omega_n + \Delta)$  is expanded as  $1 - [n\pi - l(\rho/E)^{\frac{1}{2}}(\omega_n + \Delta)]^2$ .

If  $4\Delta/\omega_n < 0.01$  solving equation (13) for  $\Delta$  gives

$$\Delta = \pm \eta\omega_n^2/2E \quad (14)$$

The frequency difference  $\delta f$  or bandwidth is  $\Delta/\pi$ , which is given by the relation

$$\delta f = \eta\omega_n^2/2\pi E = \eta n^2\pi/2\rho l^2 \quad (15)$$

This is the result given by Wegel and Walther.

From equation (14)  $\Delta/\omega = \eta\omega_n/2E \simeq 0.05$  when  $\eta^2\omega^2 = 0.008E^2$  so that  $4\Delta/\omega_n$  is not negligible compared to unity. Using the more accurate value of  $\omega_n$  given by equation (12), equation (13) becomes

$$\begin{aligned} 2\{l^2\omega_n^4\eta^2\rho/4E^3 + [n\pi - l(\rho/E)^{\frac{1}{2}}\omega_n]^2\} &= (1 + \Delta/\omega_n)^2\{l^2\eta^2\rho\omega_n^4(1 + \Delta/\omega_n)^4/4E^3 \\ &\quad + [n\pi - l(\rho/E)^{\frac{1}{2}}(\omega_n + \Delta)]^2\} \end{aligned}$$

Retaining terms in  $\Delta/\omega_n$  gives for the bandwidth

$$\begin{aligned} \delta f &= (\eta n^2\pi/2\rho l^2)(1 + 9\eta^2\omega_n^2/4E^2 - 4\Delta/\omega_n) \\ &= (\eta n^2\pi/2\rho l^2)(1 + 9\eta^2\omega_n^2/4E^2 - 2\eta\omega_n/E) \end{aligned}$$

The term  $9\eta^2\omega_n^2/4E^2$  arises from using the corrected value of  $\omega_n$  [equation (12)] and is negligible so that

$$\delta f \simeq (\eta n^2\pi/2\rho l^2)(1 - 2\eta\omega_n/E) \quad (16)$$

$$\text{If } \eta^2\omega_n^2/E^2 = 0.003, \quad 2\eta\omega_n/E \simeq 0.12$$

$$\text{and } \delta f = 0.88\eta n^2\pi/2\rho l^2$$

It will be shown later that for plastics  $\eta^2\omega_n^2/E^2$  approaches 0.003 so that Wegel and Walther's expression will be in error by about 12%. Equation (16) will give a more accurate value for  $\eta$ . Equation (16) can also be expressed in the form

$$\eta/E = (\delta f/2\pi f^2)(1 + 2\delta f/f) \quad (17)$$

#### POSITION OF NODES

The displacement at any point along the rod is given by equation (7) which reduces to

$$|\xi| = (A/\sqrt{2})[\cosh(x\omega^2\eta/E)(\rho/E)^{\frac{1}{2}} + \cos 2x\omega(\rho/E)^{\frac{1}{2}}]^{\frac{1}{2}} \quad (18)$$

when  $\eta^2\omega^2 < 0.008E^2$ .

Since  $\cosh(l\omega^2\eta/E)(\rho/E)^{\frac{1}{2}} \simeq 1$  and  $x \leq l$ ,  $\cosh(x\omega^2\eta/E)(\rho/E)^{\frac{1}{2}} = 1$  and minimum values of  $\xi$  are given when

$$2x\omega(\rho/E)^{\frac{1}{2}} = N\pi, \text{ where } N = 1, 3, 5 \dots$$



Considering a resonant frequency, i.e.  $\omega_n = (n\pi/l)(E/\rho)^{1/2}$  which gives  $x = Nl/2n = l/2$  for the fundamental. The minimum value of  $\xi$  is given by substituting for  $x$  in equation (17) retaining the second term in the expansion for  $\cosh(x\omega^2\eta/E)(\rho/E)^{1/2}$ .

Thus 
$$\xi_{\min} = \pi\omega\eta A/4E \quad (19)$$

This gives  $\xi_{\min}$  in terms of  $A$ , the displacement at the end of the rod. The minimum displacement is not zero unless  $\eta = 0$ .

Considering the fundamental frequency, it should be noted that the position of minimum displacement is at the centre ( $x = l/2$ ) only when the applied frequency coincides with the resonant frequency of the rod. Equation (12) shows that the node moves toward the free end as  $\omega$  increases because  $2x\omega(\rho/E)^{1/2} = \pi$  so that  $x$  decreases as  $\omega$  increases.

The displacement at the centre of the rod when the applied frequency is such that the amplitude at the free end is  $1/\sqrt{2}$  the maximum value when at resonance is given by putting  $\omega + \Delta$  for  $\omega$  in equation (18),  $\Delta$  being given by equation (14). Expanding the cosh and cosine terms gives

$$\xi = [A/\sqrt{(2)}] \{ (\omega^4 l^2 \eta^2 \rho / 8E) (1 + \Delta/\omega)^4 + [l(\rho/E)^{1/2}(\omega + \Delta) - \pi]^2 / 2 \}^{1/2}$$

Since  $1 + \Delta/\omega \simeq 1$  and  $\Delta = \eta\omega^2/2E$

$$\xi = \pi\omega\eta A/2\sqrt{(2)}E = \sqrt{(2)}\xi_{\min} \quad (20)$$

#### PRACTICAL CONSIDERATIONS

For most plastics and dielectrics  $E \simeq 3 \times 10^{10}$  dynes/cm<sup>2</sup> and  $\rho \simeq 1$  gm/cm<sup>3</sup>. If  $l \simeq 10$  cm., then for the fundamental resonant point  $l\omega(\rho/E)^{1/2} = \pi$ , which gives  $\omega = 5 \times 10^4$  (frequency =  $8 \times 10^3$  cycles/sec.). If  $\eta^2\omega^2/E^2 \leq 0.008$ ,  $\eta \leq 5 \times 10^4$  poise. Experiments made on a number of plastics using another method (1) indicate that at a frequency of  $2 \times 10^3$  c/s  $\eta$  lies between  $3 \times 10^4$  and  $1.3 \times 10^5$ . The method is therefore suitable for plastics although the formula given by Wegel and Walther will be in error by 12% for these values of viscosity. Equation (16) will, however, give  $\eta$  correct to about 1%. It may not be possible to determine  $\eta$  to such accuracy owing to experimental error, e.g. losses due to friction of air set in vibration and losses due to transmission of energy to the supports. The above figures differ from those given in the original paper because more accurate values of  $E$  and  $\eta$  are now available.

At higher frequencies the maximum value of  $\eta$  for which the theory is valid decreases but this is offset by the fact that  $\eta$  decreases with frequency. It appears, however, that  $\eta\omega$  is constant\* (1) so that  $\eta^2\omega^2/E^2$  is

constant (assuming that  $E$  remains constant, which is approximately true) and the theory is therefore applicable for all frequencies. The minimum amplitude, which is at the centre of the rod when at resonance in the fundamental mode, is given by equation (19) (assuming  $\eta \simeq 10^4$ ) and is  $\pi/40$  or  $0.12 \times$  amplitude at the antinode. When the frequency is such that the amplitude at the antinode is  $1/\sqrt{2}$  the amplitude at resonance, the amplitude at the centre is given by equation (19), which is  $0.17 \times$  amplitude at the antinode. It is clear that the rod should not be clamped anywhere as this will interfere with its vibration and the theory will not be valid. Some form of suspension by fine threads appears to be the most satisfactory method. Wegel and Walther advocate suspension by wires. Street (8) used fine silk threads and measured small changes in the elastic modulus of cobalt which are due to changes in the degree of magnetization of the rods.

#### CORRECTION FOR LATERAL MOTION

Equation (1) was derived on the assumption that the motion of the rod is only along its axis so that lateral forces are neglected.\* Owing to lateral expansions and contractions associated with longitudinal strain there is a small lateral motion.

Rayleigh (6) has shown from energy considerations that the effect of this motion is to increase the resonant time period in the ratio  $1/(1 + \alpha)$

where 
$$\alpha = n^2\pi^2\mu^2r^2/4l^2$$

where  $r$  is the radius of the rod and  $n$  the harmonic number. Since  $r/l$  is normally very small (of the order of 0.1),  $\alpha$  is very small. Quimby (3) considers the inertia due to lateral motion decreases the effective value of  $\mu$  (Poisson's ratio) so that the time periods are related by the equation

$$T^2/T_1^2 = E'/E = (1 + \mu')/(1 + \mu) = 1 - 2\alpha$$

since  $E = 2G(1 + \mu)$ . Thus  $G$  is altered by  $2\alpha G$  and the effect of neglecting lateral motion is to make the calculated value of the viscosity too low by an amount  $2\alpha\eta_r$  because  $G + \eta_T\partial/\partial t$  now becomes  $G + 2\alpha G + (\eta_T - 2\alpha\eta_T)\partial/\partial t$ . Since  $\alpha$  is very small the correction to  $\eta_T$  is also negligible.

#### SHEAR WAVES

In this form of tests the force is applied tangentially to one end of the rod. Considering a cylindrical tube of radius  $r$  and thickness  $\delta r$ , if  $\theta$  is the angular displacement of any section distant  $x$  from the free end  $x = 0$ , the shear strain across this section is  $r\partial\theta/\partial x$  and the

\* The correction considered here has no connection with the relation between longitudinal and tangential viscosity discussed. If there were no lateral motion ( $\mu = 0$ ) the two measures of viscosity are not equal but are related by the equation  $\eta = 4\eta_T/3$ .

\* Wegel and Walther's results also indicate that this relation is obeyed. Most of their results were for metals and glass and for these materials  $\eta$  is much less than for plastics.

elastic restoring force is  $Gr\partial\theta/\partial x$ . The viscous force is  $\eta_T r \partial^2\theta/\partial x \partial t$ . The area of the section is  $2\pi r \delta r$  so that the moment of these forces about the axis of the rod is  $2\pi r^3 G \delta r \partial\theta/\partial x$  and  $2\pi r^3 \eta_T \delta r \partial^2\theta/\partial x \partial t$ . If  $\delta x$  is the thickness of the element, the restoring couples due to the elastic and viscous forces are  $2\pi r^3 G \delta r \delta x \partial^2\theta/\partial x^2$  and  $2\pi r^3 \eta_T \delta r \delta x \partial^3\theta/\partial x^2 \partial t$  respectively. The moment of inertia of the element about the axis is  $2\pi r \delta r \delta x \rho r^2$  so that the couple due to its motion is

$$2\pi r \delta r \delta x \rho r^2 \partial^2\theta/\partial t^2$$

Equating the restoring forces to the couple due to motion gives

$$\partial^2\theta/\partial t^2 = (G/\rho) \partial^2\xi/\partial t^2 + (\eta_T/\rho) \partial^3\theta/\partial x^2 \partial t$$

The equation is identical with equation (1), the shear modulus replaces  $E$  and the tangential viscosity  $\eta_T$  now appears in place of the longitudinal viscosity. Comparison of  $\eta$  and  $\eta_T$  obtained from the two forms of test should show the relation

$$\eta = 4\eta_T(1 + \mu)^2/3$$

to hold.

Wegel and Walther find this relation is satisfied in some cases but in others the longitudinal viscosity was too high which was taken to indicate that dissipation occurs in uniform dilatation, i.e. the existence of a volume viscosity  $\eta_v$ . Frenkel and Obrastzov (9) also make this supposition but no serious experimental work has yet been undertaken to investigate this. It is proposed to measure  $\eta$  in torsion and by longitudinal waves. The torsion tests will give  $\eta_T$  directly whereas longitudinal tests give  $\eta$ . If  $\eta > 4(1 + \mu)^2\eta_T/3$  a volume viscosity exists which can be calculated from the expression given earlier.

## CONCLUSIONS

The coefficient of internal friction of materials can be calculated by studying the resonance curves of rods when subjected to mechanically alternating stress. The correct relation between the longitudinal and tangential coefficients of friction is also deduced. If a volume viscosity exists, an expression is derived which enables the coefficient to be determined experimentally. It is important that the specimen should not be rigidly clamped because there is no true node if friction is present. Suspension by light threads near the nodal points is recommended.

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# The Application of Multiple Factor Analysis to Industrial Test Data

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The technique known as Multiple Factor Analysis is outlined and its relevance to problems of industrial physics is discussed. In particular, the method is applied to the analysis of a set of rheological and electrical tests on a set of plastics. Three "factors" are sufficient to account for all the observed relationships between the ten tests. Three numerical solutions have been evaluated, one involving non-orthogonal "factors" presenting the simplest solution for interpretation. All the solutions are linear transformations of one another. (See also the following paper.)

## STATEMENT OF THE PROBLEM

In many industrial and research problems measurements are made on a variety of samples of materials in a comparatively large number of tests. These tests range from precise measurements of physical properties to standardized tests of a purely empirical nature. Considered as sets of measurements the data are frequently too extensive to permit understanding and interpretation. Some method is therefore necessary for the efficient condensation of the data and the extraction of any fundamental relationships. This is precisely the problem which has faced the psychologists in relating and assessing the structure of mental tests. For this purpose the techniques of Factorial Analysis have been developed by mathematical psychologists, in particular Burt (1), Thomson (2) and Thurstone (3), who have suggested that useful applications might be found outside the psychological sphere. The techniques are not necessarily in their final form nor are all the mathematical problems fully worked out. This, however, does not preclude useful application, and no doubt many incompletely developed sections might receive useful contributions to their study as a result of wider dissemination.

The problems which are treated by Factor Analysis are rather different from those usually dealt with by other statistical methods in that the aim is not to study treatments which produce significant effects and interactions, but rather to endeavour to isolate the most effective means of characterizing a set of materials (or persons) which differ in their measurement in a variety of tests and to study the relationships between the tests used. Such factorial studies lead to the design of new tests and the carrying out of further planned experiments. A preliminary application of Factor Analysis to study the relationships between a number of rheological tests on Cheddar cheese has recently been reported and the results used in planning a new experiment (4). In order to give further information as to the value of the technique of Factor Analysis as applied to such problems the present data, representing rheological and electrical tests on a group of plastics, were selected for a systematic study. It should be emphasized that the data were not

specially designed for a factorial study but represent information which was already available. The only condition for inclusion was that every sample of material utilized should have been measured in each test.

## GENERAL OUTLINE OF FACTOR ANALYSIS

There are in existence a number of slightly different forms of analysis, but these are to be regarded as various approximations and computational compromises towards a "least square" solution of the problem. The first step is the formation of a matrix of correlation coefficients representing the intercorrelation of each pair of tests. The diagonal entry is left blank since it is not part of the experimentally determined data. The aim of the analysis is to extract a limited number of factors in terms of which the observed intercorrelations may be reproduced, within the limits of experimental error, according to the following equation

$$r_{ab} = a_1b_1 + a_2b_2 + \dots \quad (1)$$

where  $a_1$  and  $b_1$  are the "factor loadings" of tests  $a$  and  $b$  in Factor I, and  $a_2$  and  $b_2$  the corresponding loadings in Factor II, etc. The factors as usually extracted are orthogonal, i.e. the sets of factor measurements and factor loadings are uncorrelated. The reader must refer to one of the general texts for the precise details of the computational techniques. If, as is usually the case, the analysis is restricted to "common factors" (those involved in more than one test) the appropriate entry in the diagonal cells of the original matrix is the communality ( $h^2$ ) for each test defined as follows

$$h_a^2 = a_1^2 + a_2^2 + \dots \quad (2)$$

In any particular case the communality has to be specified in relation to the number of factors on which it is based but, in the first instance, it has to be estimated and a process of successive approximation carried out. The communality represents the proportion of the variance in each set of test measurements accounted for by the "common factors" extracted by the analysis. The factors represent convenient reference axes by means of which the relationship between the tests may be expressed

and, considering a "factor" as an ideal test which may be designed or be approximated to by the statistical combination of several tests, then the factor loadings represent the correlation between such reference factors and the actual tests.

Provided there are no more than three common factors, then, using the factor loadings as co-ordinates, it is possible to construct by means of two-dimensional or three-dimensional models the pattern of relationships between the various tests. For more than three factors recourse is necessary to the formal mathematics of the geometry of hyper-space, but useful interpretation can be obtained by a series of three-dimensional models or by plotting the factorial solution, two axes at a time. In the present application it has not been necessary to go beyond three dimensions for the differences between the original correlations and those reconstructed from three factors are statistically insignificant. The quantity  $(1 - k^2)$ , often termed the "uniqueness," represents the proportion of the variance of the measurements in a particular test which cannot be accounted for by the common factors. This may also be termed the specific variance. The reasons for a high value of the specific variance may be:

- (1) The failure to include in the group of tests another related to the one found to have a "high specific." That is, the test in question samples aspects of the behaviour of the materials (or persons) not covered by any other test;
- (2) The existence of non-linear relationship between certain tests;
- (3) The existence of a high degree of random error in the measurements obtained by means of the test concerned.

If the errors of measurement are known, in the form of test-retest correlations, it is possible to separate the effects of specific variance and error variance.

From this preliminary discussion it should be clear that a factor may be a common factor, a group factor or a specific factor depending on the particular set of tests which are analysed. The reference axes are essentially the product of the accident of selecting one particular group of tests and materials rather than another. Furthermore, the same set of observed relationships may be reproduced by co-ordinate loadings referred to an infinite number of sets of orthogonal axes or, for that matter, sets of oblique axes. One of the central problems of factor analysis is how to select and interpret the most meaningful set of axes. In this guidance has to be taken, in the main, from scientific intuition together with any obvious relationships derived from the data themselves. Attempts have been made to develop purely statistical criteria for determining the most meaningful set of axes but at present no final procedure has been developed. The factors have to be interpreted by relating them to the pattern of factor loadings in the various tests. This process will later be illustrated by the actual data. To

summarize, the technique of the Factor Analysis starting with a matrix of correlation coefficients between the tests thus:

Test	a	b	Test d	.	.	n
a	—					
b	$r_{ab}$	—				
c	$r_{ac}$	$r_{bc}$	—			
d	$r_{ad}$	$r_{bd}$	$r_{cd}$	—		
.	.	.	.	.	—	
.	.	.	.	.	.	—
n	$r_{an}$	$r_{bn}$	$r_{cn}$	$r_{dn}$	.	.

reduces this to a set of factor loadings involving a smaller number of factors than the original tests

Test	I	Factor II	III
a	$a_1$	$a_2$	$a_3$
b	$b_1$	$b_2$	$b_3$
c	$c_1$	$c_2$	$c_3$
d	$d_1$	$d_2$	$d_3$
.	.	.	.
.	.	.	.
n	$n_1$	$n_2$	$n_3$

such that

$$r_{ab} = a_1b_1 + a_2b_2 + a_3b_3 + \dots$$

The initial solution may give axes which are determined essentially by statistical accidents of sampling and rotation of axes may be necessary before a clear interpretation of a solution is possible.

#### EXPERIMENTAL DATA

The materials selected for the tests were various polyvinyl-chloride-plasticizer mixes taken from experimental and production materials over a period of some two years. A large number of materials had been tested but only those for which a complete set of measurements were available are considered in this work. At least four different grades of polymer were in use during this period. These were made up with various proportions of one or more of eight plasticizers and, in addition, contained small quantities of various fillers and stabilizers. Measurements thus cover a fairly broad range of materials with diverse compositions and properties. Seven of the tests were rheological and three were electrical. The tests will be denoted by letters, as follows:

A. Tensile strength measured at a standard rate of extension at 20° C. Average of five results taken;

B. Percentage elongation at break measured at the same time as the tensile strength. Average of five results taken;

C. Percentage elongation in five minutes under a dead load adjusted to the same initial stress at 20° C. Average of two results taken;

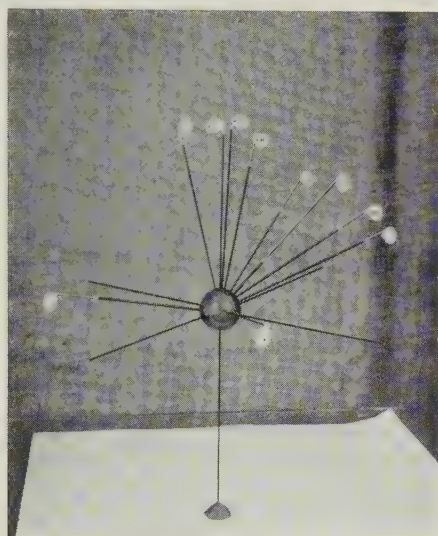
D. Percentage of the elongation in test C recovered in five minutes after removing the load. Average of two results taken;



Table 1. *Correlation matrix*

Test	- A	+ B	+ C	+ D	+ E	+ F	+ G	- H	+ I	- J
- A	—									
+ B	0.684	—								
+ C	0.812	0.657	—							
+ D	0.216	0.258	0.315	—						
+ E	0.843	0.659	0.966	0.352	—					
+ F	0.729	0.518	0.874	0.301	0.897	—				
+ G	0.539	0.532	0.421	0.278	0.378	0.315	—			
- H	0.631	0.503	0.534	0.256	0.549	0.445	0.567	—		
+ I	0.544	0.611	0.519	0.242	0.530	0.345	0.535	0.714	—	
- J	0.096	0.084	0.214	- 0.070	0.111	0.204	0.281	- 0.163	- 0.161	—

E. B.S. Hardness Number defined by the indentation in 30 sec. produced by a standard ball under defined conditions. Average of four measurements taken;



Three-dimensional analysis of data (the termini of the test vectors are represented by the small white beads and should be distinguished from the labels which are set at equal distances—unity—from the origin)

F. The percentage indentation in 24 hr. at a temperature of 70° C. by a loaded wire passing over the material bent over a 2 in. diameter steel rod. Average of two measurements taken;

G. *Minus* the temperature in deg. C. to which a set of four samples must be lowered before two of them break when bent in a standard way;

H. The logarithm of the volume resistivity. One reading only;

I. Dielectric constant measured at 800 c/s on the same sample as H. One reading only;

J. Power factor measured at 800 c/s on the same specimen as H and I. One reading only.

In actually working out the correlations the measurements were multiplied, as required, by constant factors

or referred to an arbitrary origin. These procedures have no effect on the correlations themselves but reduce considerably the labour involved in computation. The correlation matrix is reproduced in Table 1 with the signs of the appropriate tests reversed so as to make the entries substantially positive. This is the first stage in the analysis by Thurstone's Simplified Centroid Method (5) which uses estimated values for the diagonal cells. The first solution was then used to give a better approximation for the diagonal entries and the analysis repeated. As Thurstone has himself criticized the Simplified Centroid Method, the second approximation was used for a further two approximations by his Complete Centroid Method, see Thurstone (3), p. 161. In the earlier stages of the analysis the entries in the diagonal cells were adjusted as required. In later approximations the calculations were performed without alteration to the original diagonal entries or the residuals, at least as far as the extraction of a fourth factor. The differences between the third and fourth approximations were small, so that the solution given by the fourth approximation was regarded as stable and is reproduced in Table 2.

Table 2. *Factor matrix (unrotated)*

Test	Factor			
	I	II	III	$h^2$
- A	+ 0.86	+ 0.10	+ 0.07	0.75
+ B	+ 0.75	+ 0.03	- 0.10	0.58
+ C	+ 0.92	+ 0.14	+ 0.30	0.95
+ D	+ 0.35	+ 0.08	- 0.11	0.14
+ E	+ 0.92	+ 0.27	+ 0.29	1.00
+ F	+ 0.81	+ 0.16	+ 0.42	0.85
+ G	+ 0.71	- 0.57	- 0.41	1.00
- H	+ 0.69	+ 0.17	- 0.36	0.63
+ I	+ 0.68	+ 0.19	- 0.50	0.76
- J	+ 0.17	- 0.58	+ 0.41	0.53
$\Sigma k^2$	5.22	0.87	1.09	7.18
$\frac{\Sigma k^2}{n}$	0.52	0.09	0.11	0.72

At the border of this table the sum of the squares of the factor loadings are given, these indicating the

proportion of the variance accounted for by the common factors extracted by the analysis. The sum of the squares of the entries in any one row, which gives the communality of a test, represents the proportion of the variance of the measurements in that test accounted for by the common factors. The sum of the squares of the entries in any column, expressed as a proportion of the number of tests, gives the proportion of the total variance accounted for by that particular factor. The actual data quoted are given to two places of decimals although four were used throughout in the calculations; to retain more than two at this final stage would give an impression of spurious accuracy. The unrotated solution will not be discussed in detail for, as has already been indicated, it does not necessarily represent the most meaningful solution, i.e. the one which most effectively shows up the relationship between the tests and lends itself to a most satisfactory interpretation. The amount of the variance of the measurements in each test accounted for by the common factors will, however, be unchanged by rotation so that the communalities can be interpreted forthwith. The main points to note are:

(1) That the three common factors account for 72% of the total variance;

(2) Tests C, E and G have extremely high communalities, approaching unity. Therefore the errors of measurement in these tests must be small. Recalling the nature of test G—a fracture test—this is surprising;

(3) Test D has a very low communality. Therefore either test D involves a large error of measurement or measures some "property" of the materials not covered by the remaining tests. As D is the only test involving springiness or recovery of the material, it is possible that both conditions hold. The analysis, however, stresses the importance of test D, the need for investigating the random errors of measurement and for the development of new tests of springiness so that a common factor of this origin may be isolated by the inclusion of more than one test involving springiness in a new analysis.

(4) Since the electrical tests are believed to be fairly accurate (reproducible), the moderate communality for these tests suggests that either each involves properties which are not involved in the remaining tests or, equally likely, the relationships between these and some of the other tests may be non-linear (for the particular samples tested). This point requires further investigation.

(5) Tests A and B are both fracture tests and, as such, it is to be expected that there would be considerable error of measurement. Since these tests are destructive, only the variation between samples can be taken as an estimate of error. On the basis of the data so far studied it is not possible to state whether the moderate communality of these two tests is due to error, non-linear relationships to other tests or the fact that each test samples some specific property in addition to common factors. These points, also, require further investigation.

## ORTHOGONAL ROTATION

Two sets of orthogonal axes have been derived from the original solution. In the first set rotation was performed, taking two axes at a time, to make the number of negligible factor loadings a maximum. This analysis is given in Table 3. From the main features of the pattern of factor loadings in relation to the qualitative nature of the tests and their hypothetical relationships any inference as to the nature of the factors has to be made. We do not, however, know in advance whether an orthogonal set of axes will provide the most satisfactory interpretation of the data or whether recourse will have to be made to oblique axes. Nor do we know if it will be possible to identify any of the factors with specific physical properties. In the present solution, Factor I', which accounts for 48.4% of the total variance, has almost constant loadings in tests — A, B, C, E, G, — H and I with a negligible loading in test — J. What could be the essential nature of any "property" correlating equally with measures of tensile strength, stretching under constant load, penetration with a ball indenter, brittleness as defined by test G, dielectric constant and volume resistivity? Something of the nature of the molecular cohesion of the material suggests itself. Factor II' suggests no clear interpretation, but Factor III' represents predominantly a link between tests G and — J. Since this is more effectively demonstrated by a later analysis, comments will for the time be withheld.

Table 3. *Orthogonal rotation (maximizing the number of negligible factor loadings)*

Test	I'	Factor II'	III'	
— A	+ 0.80	+ 0.34	+ 0.04	
+ B	+ 0.75	+ 0.13	+ 0.03	
+ C	+ 0.78	+ 0.58	+ 0.07	
+ D	+ 0.37	+ 0.02	— 0.07	
+ E	+ 0.79	+ 0.60	— 0.05	
+ F	+ 0.64	+ 0.66	+ 0.08	
+ G	+ 0.78	— 0.36	+ 0.51	
— H	+ 0.77	— 0.08	— 0.18	
+ I	+ 0.81	— 0.20	— 0.24	
— J	+ 0.01	+ 0.23	+ 0.69	
$\Sigma k^2$	4.84	1.48	0.86	$\Sigma \Sigma k^2$ 7.18

A second orthogonal rotation of the original factor axes was carried out so as to make one axis pass through the B.S. hardness test which, having a communality of almost unity, is adequate to define a factor. The solution is given in Table 4. Factor I'' accounts for 47.5% of the total variance in all the test measurements. At the time of writing no precise criterion had been worked out for the error of a single entry in the table of factor loadings. For discussion purposes, factor loadings not exceeding 0.20 will be deemed negligible. On the basis of this rough criterion, factor II'' represents, in the main, a link between tests G and — J. The resemblance of



factor II'' to factor III' of the previous rotation is striking. Factor III'' represents primarily a link between tests G, — H and I.

Table 4. Orthogonal rotation making B.S. Hardness Number Factor I''

Test	I''	Factor II''	III''	
— A	+ 0.83	— 0.09	— 0.22	
+ B	+ 0.67	— 0.08	— 0.35	
+ C	+ 0.97	— 0.12	— 0.01	
+ D	+ 0.31	+ 0.04	— 0.21	
+ E	+ 1.00	+ 0.00	— 0.01	
+ F	+ 0.90	— 0.12	+ 0.13	
+ G	+ 0.37	— 0.56	— 0.74	
— H	+ 0.57	+ 0.13	— 0.54	
+ I	+ 0.53	+ 0.19	— 0.66	
— J	+ 0.11	— 0.69	+ 0.20	
$\Sigma k^2$	4.75	0.89	1.54	$\Sigma \Sigma k^2$ 7.18

#### NON-ORTHOGONAL ROTATION

If the original three-factor solution is used to construct a solid model then several striking features which can only be inferred from the two-dimensional analysis become evident. The lines drawn from the origin to the points defined by the use of the separate factor loadings as co-ordinates are usually termed the test vectors and the length of each vector is equal to the square root of the communality. The correlation between two tests is given by the expression

$$r_{ab} = h_a h_b \cos \phi_{ab} \quad (3)$$

where  $h_a$  and  $h_b$  are the lengths of the test vectors  $a$  and  $b$  and  $\phi_{ab}$  the angle between them. The three-dimensional analysis of the present data given in the figure suggested that the termini of all the test vectors except D and — J lie in a single plane not passing through the origin. Secondly, tests — A, B, C and F form one plane and test E, D, — H and I form a second plane at a small angle, both passing through the origin. Possibly these two planes define a single plane, within the limits of error. The vectors I, G and — J form approximately an independent plane, but the number of points is insufficient to determine this plane very precisely.

Reference axes may be selected perpendicular to these planes so that the structure in the data may be effectively seen from the pattern of non-orthogonal factor loadings. The question arises whether such reference axes have any special interpretation or whether the test vectors at the intersection of the planes defined by the data have any special significance. (In the sphere of mental testing the vectors at the intersection of these structural planes have been suggested by Thurstone as defining primary mental abilities.)

One set of non-orthogonal reference axes has been derived by numerical processes in such a way as to follow most closely any structure shown by the data. The procedure used was that described by Thurstone (3, p. 194),

and the fourth approximation is given in Table 5 together with the direction cosines of the reference axes representing their mutual intercorrelation. To distinguish

Table 5. Details of non-orthogonal rotation: (a) non-orthogonal rotation of axes; (b) correlation between the reference vectors; (c) transformation matrix  $\Lambda_{01}$

( $V_1 = F_0 \Lambda_{01}$ ,  $V_1$  represents the non-orthogonal factor matrix,  $F_0$  the original factor matrix,  $\Lambda_{01}$  the transformation matrix.)

	Test	Factor		
		$\alpha$	$\beta$	$\gamma$
(a)	— A	+ 0.79	+ 0.06	+ 0.54
	+ B	+ 0.74	+ 0.10	+ 0.35
	+ C	+ 0.78	+ 0.03	+ 0.76
	+ D	+ 0.37	— 0.02	+ 0.10
	+ E	+ 0.80	— 0.09	+ 0.74
	+ F	+ 0.64	— 0.00	+ 0.79
	+ G	+ 0.72	+ 0.68	+ 0.12
	— H	+ 0.78	— 0.06	+ 0.09
	+ I	+ 0.83	— 0.08	— 0.03
	— J	— 0.05	+ 0.61	+ 0.47
(b)	$\alpha$	1.000		
	$\beta$	+ 0.030	1.000	
	$\gamma$	+ 0.267	+ 0.204	1.000
(c)	I	+ 0.938	+ 0.178	+ 0.575
	II	+ 0.129	— 0.983	+ 0.078
	III	— 0.322	+ 0.031	+ 0.815

these axes from the orthogonal sets they have been denoted by  $\alpha$ ,  $\beta$  and  $\gamma$ . Although the axes have been permitted freely to depart from the orthogonal position, axes  $\alpha$  and  $\beta$  remain almost at right angles whilst  $\alpha$  and  $\gamma$  and  $\beta$  and  $\gamma$  are substantially correlated. In addition, all the loadings become positive or negligible. The special features of this solution are:

(1) Factor  $\alpha$  plays no part in the variance of the measurements in test J;

(2) Apart from test D, whose special significance has been discussed earlier, the differences between the remaining loadings in factor  $\alpha$  are small, probably insignificant. The resemblance between factor  $\alpha$  of the present analysis and factor I' of the first rotation are striking and lead to the suggestion that factor  $\alpha$  may define a fundamental property of the material. It is of special interest to note that the measurements of dielectric constant and volume resistivity correlate to a high degree with such a postulated property and that these two tests have no component represented by factors  $\beta$  and  $\gamma$ ;

(3) Factor  $\beta$  represents now a pure doublet, that is, a relationship between only two tests. Its resemblance to factor III' and II'' is obvious;

(4) Factor  $\gamma$  is not easy to interpret but, when the insignificant loadings are deleted, factor  $\gamma$  represents a link between the indentation or extension tests and the

power factor. The three tests involving steady deformation or extension have almost identical loadings.

(5) Factors  $\alpha$ ,  $\beta$  and  $\gamma$  form the most effective set for the representation of the structure of the data and may be easier to interpret as a whole than the orthogonal sets. These reference axes are not, however, identical with the primary vectors formed by the intersection of the structural planes determined by Thurstone's method (3, p. 353). If the corresponding loadings with respect to the primary vectors are to be calculated, then a further transformation is necessary to a set of axes orthogonal in pairs to the three reference axes  $\alpha$ ,  $\beta$  and  $\gamma$ . The special property of the primary vectors in relation to the non-orthogonal reference frame  $\alpha$ ,  $\beta$  and  $\gamma$  is that they have unit complexity which may be represented by the following table of direction cosines:

	$\alpha$	$\beta$	$\gamma$
$T_1$	$d_1$	0	0
$T_2$	0	$d_2$	0
$T_3$	0	0	$d_3$

Where  $T_1$ ,  $T_2$  and  $T_3$  represent the primary vectors

and  $d_1$ ,  $d_2$  and  $d_3$  are constants (less than or equal to unity).

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## Note on the Interpretation of Multiple Factor Analysis

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[Paper received 7 March, 1949]

The Multiple Factor Analysis of the preceding paper shows that the tests described therein each measure some combination of three fundamental factors, each factor representing a property or quasi-property of the material. The present note indicates one way in which this knowledge may be developed, the vector model being interpreted in terms of current ideas on high-polymer molecules. These lead to considerations of the grouping of the vectors of the vector model which indicate that the two most important factors may be the freedom of rotation of the carbon-carbon polymer chain linkages and the polar nature of the plasticizers. This conception is shown to agree with the whole structure of the vector model.

In the preceding paper a general picture has been given of the points that arise from the consideration of the statistically derived vector model when each of four sets of factor axes are used. In that paper the authors have derived from the experimental data as much information as can at present be obtained by the methods of Multiple Factor Analysis. This information was obtained by consideration of a vector model derived by a purely numerical process from the original data only and no other experimental or theoretical information was used. It is clear that the methods of Multiple Factor Analysis, although giving immediate valuable information about the numerical connexion between tests, will not give directly any pictures of the physical processes involved in the tests. Their main use is to systematize the testing of the materials under consideration by eliminating unnecessary, and selecting desirable, test methods. An

interpretation of the vector model is, however, interesting, and that given below is based on generally accepted theories of the behaviour of plasticized high polymers (1). It must be realized, however, that the matter of interpretation is controversial so that these suggestions should be considered as tentative. The position of the axes in the original vector model, as explained in the paper by Harper, Kent and Scott Blair, is fortuitous. The first orthogonal rotation is of theoretical interest but the most useful and meaningful sets of axes are the second orthogonal rotation and the non-orthogonal rotation. Tentative interpretations will therefore be confined to the latter two.

The second rotation was performed so as to make the B.S. Hardness Test Vector coincident with a factor axis, namely Factor I''. Thus Factor I'' is B.S. Hardness and it is noticed that this factor is important in all the tests



except test — J. Tests E, C and F are all measures of hardness, being measures of the deformation in a fixed time under a given stress (hardness is here considered as a property or quasi-property approximating to that measured by the B.S. Hardness Test). This deformation is the sum of deformations due to three causes, namely change of intermolecular distances under stress, high elastic deformation and viscous deformation. The dominant deformation is that due to high elasticity, so tests E, C and F (whose vectors are closely grouped) may be considered to be tests of high elastic properties.

In high elastic deformation of the polyvinyl chloride compositions which are the subject of the data, randomly-coiled polymer chains change their configuration to a less probable one, the rate of change depending upon the freedom with which the segments can rotate about the carbon-carbon linkages. As the configuration can be modified by the rotation of one or more units (up to a limit set by chain entanglements) there will be a distribution of orientation times covering the various mechanisms. The freedom of movement of the segments will depend upon the plasticizer content and, to some extent, the polymer-polymer and polymer-plasticizer dipole coupling. The dielectric constant depends upon contributions of both the polymer and plasticizers. Considering firstly the polymer, polyvinyl chloride, the dipole is carbon-chlorine, so that for dipole orientation to occur there must be rotation about the carbon-carbon linkages. Thus the dielectric-constant test depends upon the same carbon-carbon link rotation as do the high-elastic properties of tests E, C and F. However, this electrical test will also depend to some extent upon the dipole moment of the plasticizers. Since the dielectric constant (test I) depends upon Factors I'' and III'' Factor III'' can be tentatively identified as a factor related to the polar nature of the plasticizers. Factor II'' has been seen to be mainly a link between tests — J and G. No attempt to interpret it will be made in the present note.

Having given tentative labels to two of the factors it can now be seen to what extent they will explain the structure of the vector model. Vectors E, C and F, as has been seen, approximate to Factor I''. Tests H and I which depend upon both Factors I'' and III'' are placed between these. The above fit can be expected as it was upon these tests that the tentative identification of axes was based. It is the fit of the other tests which is important. Tests A and B are tensile failure tests. There is reason (2) to believe that the failures occur by the following steps: firstly, uncoiling and alignment of the polymer chains against the forces of high elasticity, involving rotation about the C-C linkages (involving the forces of rotational hindrance of Factor I'') and secondly, movement of the chains past one another just prior to and when failure takes place. The forces involved in this second phase will largely arise from the van der Waals attractions between chains which will be modified by the amount of plasticizer and its polar properties.

Thus, vectors A and B lie between axes I'' and III'' but nearer the former. Test D will measure, besides viscous flow (if any), the extent to which the material fails to recover in the time of the test. This is governed by the "distribution of orientation times" which is an expression of Factor I''. If there are any new interchain linkages formed during the experiment, such as the formation of crystallites, high elastic deformations will not completely recover. The formation of these linkages depends upon the compatibility of the plasticizers with the polymer which is to some extent dependent upon the polar properties of the plasticizers (Factor III''). Thus vector D lies midway between the axes I'' and III''.

Test G is a fracture test, so that it should be expected to have connexions with tests — A and B. Since, however, it is performed more rapidly than — A and B, and at a much lower temperature, the connexion will not be too close. The molecular processes involved in test G, which takes about 1 sec., would be very rapid at room temperature. They are likely to compare to some extent, therefore, with the electrical a.c. tests (these a.c. tests involve orientation times of the order of  $10^{-3}$  sec., whereas most of the rheological tests require 30–300 sec. to perform). Thus vector G lies in the region between the tensile failure vectors — A and B and the electrical vectors I and — J. Thus this interpretation of the two Factors I'' and III'' fits and explains the positions of all the test vectors except — J, which vector depends upon Factor II'' which has not been interpreted. The extension to non-orthogonal axes is of considerable theoretical interest, but it appears unlikely that, from the purely practical outlook, their use would provide much more information from this particular set of data. When non-orthogonal factors are used the picture obtained from orthogonal factors may be modified on the following lines.

Factor  $\alpha$ , as has been seen, has a heavy (and substantially equal) loading in every test except test J. It is noticed that test I is an almost perfect measure of Factor  $\alpha$ , its loadings in Factors  $\beta$  and  $\gamma$  being negligible. Therefore  $\alpha$  may be considered as the dielectric properties of the material (previously the dielectric constant was being split into two parts, due respectively to the polymer and the plasticizer, but now the material should be considered as a whole). Factor  $\gamma$  chiefly represents the three tests C, E and F which are the three tests in which the deformation in a given time under a steady load is measured. This factor is most probably hardness, but not quite the hardness measured by test E. Factor  $\beta$  as has been seen, is purely a link between tests G and — J; as in the case of Factor II'' no attempt to interpret it will be made.

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## NOTES AND NEWS

## New Books

**Scientific Foundations of Vacuum Technique.** By SAUL DUSHMAN, Ph.D. (New York: John Wiley and Sons, Inc.; London: Chapman and Hall, Ltd.) Pp. xi + 882. Price 120s. net.

There can be no doubt that this treatise is the largest, most comprehensive and up-to-date work on the subject of vacuum technique. Dr. Dushman is an acknowledged expert in this field and such a volume from his pen is most welcome. He was interested in the subject before the introduction of the diffusion pump; he saw its invention by Gaede and was associated with Langmuir during its development. As a result of his long experience, therefore, he is singularly well fitted for writing such a book, now that vacuum technique has emerged from the laboratory into the engineering stage of practical application.

The scope of the book can be realized from a survey of the twelve chapter headings, viz.: Kinetic Theory of Gases; Flow of Gases through Tubes and Orifices; Mechanical Pumps; Steam Jet Ejectors and Mercury-Vapour Pumps; Vapour Pumps using Organic Liquids; Manometers for Low Gas Pressures; Sorption of Gases and Vapours by Solids; Sorption of Gases by "Active" Charcoal, Silicates (including Glasses), and Cellulose; Gases and Metals; Chemical and Electrical Clean-up of Gases at Low Pressures; Vapour Pressures and Rates of Evaporation; Dissociation Pressures of Oxides, Hydrides and Nitrides, and Rates of Oxidation. From this list of contents it can be seen that, as might be expected, the emphasis is on the discussion of sealed-off rather than continuously-evacuated equipment; as the title implies, the basis has been the fundamental ideas underlying the subject rather than its more practical aspects. Dr. Dushman has treated the subject from very many points of view and the book is a veritable mine of information.

If such an outstanding book could be criticized, it would be to a great extent from the point of view of its editing. Perhaps the most unusual feature is the large amount of verbatim transcription from original papers, which is printed in small type; this tends to break up the continuity and suggests that the book is more a collection of pieces of information rather than a balanced and reasoned exposition of the subject such as would be expected in a book of this type. Possibly it is because the scope has been made too large. For example, the two chapters on Sorption alone take 160 pages and, in fact, comprise in themselves a text-book so detailed that the ordinary vacuum engineer might be forgiven for losing track of the main features. Again, the treatment in the chapter on gases and metals is more that of an encyclopaedia than a readable survey. The section on commercial rotary pumps, rightly or wrongly, is nothing more than a collection of data obtained from manufacturers' catalogues and faithfully reproduces their idiosyncrasies without any attempt at reducing them to a common denominator. It is scarcely conducive to easy reference to find under the chapter-heading "Mechanical Pumps" an historical review of the birth of the oil diffusion pump, a discussion of the speed of a pump (obviously largely written with the diffusion pump in mind), the elements of the design of a high vacuum system and the determination of the speed of a

pump, again partly with special reference to the diffusion pump. Similarly, several non-fractionating oil diffusion pumps are described under the general heading "Fractionating Types of Oil-Vapour Pumps." It might be noticed that the symbol  $Q_{ul}$  has been used to denote the number of micron litres per second, whereas  $P_{\mu b}$  refers to the pressure in microbars.

No reference has been made to the improved high-speed single-stage diffusion pumps mentioned by Gaede in his 1923 paper; this leaves the impression that Gaede's diffusion pump has inherently a low speed.

In spite of these, in the reviewer's opinion, small defects, Dr. Dushman's book is fundamentally sound and can be recommended to anyone wishing to become conversant with the scientific foundations of the technique. The book has been beautifully produced and is profusely illustrated with line diagrams. The proof-reading has been extremely thorough, only two small printing errors having been noticed. Dr. Dushman is to be congratulated on what is likely to be a standard work for many years to come. R. WITTY

**The Fundamentals of Gas Turbine Technology.** By W. R. THOMSON, B.Sc.(Tech.), A.M.Inst.C.E. (London: Power Jets Ltd.) Pp. 148. Price 25s. net.

In a time of careless titles the exactness of the name of the present book is pleasing. It might so easily have been called *The Fundamentals of Gas Turbines*, which it is not. It is indeed an excellent work on its given title and gives a very clear appreciation of the fundamental material and procedures in this particular technology. More than that, however, in the way in which the author has accomplished his task, he provides a model example of how to set down the fundamental material and procedures in any other limited technology. The freedom which has presumably been accorded by the publishers, Power Jets Research and Development Ltd., should be much appreciated, as it is not often that a private undertaking publishes widely, and to such an extent, procedures and concepts which its own staff have played so great a part in developing.

The physicist would probably like to see more discussion of the fundamental physics than are given herein but, on the other hand, the author in his title has clearly told him not to expect this. The mechanical engineer may be disappointed to find only very little discussion of comparisons between gas-turbine thermodynamic cycles and those already more familiar to him. He will find compensation, however, in the clarity of exposition of the various cycles which are discussed and will indeed find the book invaluable if he is entering this new field. From the physicist's point of view, again, it is worth remarking that the author gives a much clearer and fuller discussion of the heat-engine cycles with which he deals than is usually found in heat-engine text-books. This is presumably due to the need which he and his colleagues have experienced for clarifying their concepts in this rapidly developing field where existing mechanical engineering practice offered little guidance. One point which one might query is the peculiar statement on p. 19 that "Entropy is truly a mathematical and not a physical property of the medium."

The discussion of the behaviour of flow velocity in nozzles



throughout the range from subsonic to supersonic speeds is exceedingly good and will be helpful to many mechanical engineers not concerned with gas turbines. One regrettable omission, however, is the lack of extensive discussion of the heat exchange problems which are fundamental in the development of efficient gas turbine installations.

The many diagrams throughout the book are exceedingly clear and the production is excellent. Only a few typographical errors have been noted and none of these is in an important formulae.

R. S. SILVER

**Reports on Progress in Physics.** Edited by A. C. STICKLAND, Ph.D. Vol. XII (1948-49). (London: The Physical Society.) Pp. 382. Price 42s. net.

The annual volume of *Reports on Progress in Physics*, published by the Physical Society, is now a well-established and, in fact, an indispensable part of scientific literature not to be ignored by any physicist who hopes to keep himself in touch with the manifold developments of his subject. No more is really required of a reviewer than to call the attention of any reader who may have missed the announcement to the fact that Volume XII (1948-49) is now available and that it is standard does not fall below that of its predecessors. The contents include articles on: Mass Spectrometry, Nuclear Paramagnetism, Phosphors and Phosphorescence, Spontaneous Fluctuations, Nuclear Experiments with High-Voltage  $\alpha$ -Rays, Linear Accelerators, Viscosity in Glass, Theory of Oxidation in Metals, Fracture and Strength of Solids, Multi-pole Radiation, Collisions between Atoms and Molecules, Low Temperature Physics, Slow Neutron Absorption Cross-Sections of the Elements, and Molecular Distribution and the Equation of State in Gases—a well-chosen and representative selection from among the more active sections of Physics. The authors are all acknowledged specialists on the subjects on which they report and many of them have mastered the difficult art of presenting a readable and not too incomprehensible review of their speciality to their fellow physicists. Paper, printing and binding are, by present-day standards, excellent.

Any attempt to appraise the individual items in this very varied assortment would be unfair to their authors since it could only reflect the personal tastes or interests of the reviewer. A physicist, whose "mathematical ceiling" approximates more to that of a cottage than a cathedral, may, for example, well find some of the more highly mathematical contributions very heavy going; but this is no disparagement either of their importance or of the skill with which they are presented. It is not every mathematician who possesses the art, so admirably displayed by Professor Mott in his joint contribution to the present volume, of making an abstruse subject both fascinating and apparently comprehensible to the non-specialist. Perhaps, some day, the Physical Society will solve the difficulty by presenting us with a companion series of Reports on Progress in Theoretical Physics.

One may, however, make the comment that, on the whole, the shorter reports seem to present a more satisfactory picture of their subject than those of greater length: it is possible to plumb the outlines of a sketch by too much attention to detail. The admirable lists of references at the end of each report tell the reader where he may look for more if he requires it. One may also express one's gratitude to those among the present authors who have adopted Lord Rutherford's dictum: "Whatever your audience, and however deeply you mean to go into your subject, always begin at the beginning." One

would, however, like, if it is not indiscreet, to ask the computers of the table of neutron absorptions, "What is a barn?" But perhaps this information has not yet undergone "declassification."

J. A. CROWTHER

**Industrial High Frequency Electric Power.** By E. MAY, B.Sc., M.I.E.E. (London: Chapman and Hall, Ltd.) Pp. xi + 355. Price 32s. net.

This book deals with the generation of high-frequency electric power and with its use in industry for induction and dielectric heating processes. An introductory chapter summarizes the basic theory of oscillatory and coupled circuits.

The h.f. power generators described include arc and spark-gap oscillators, mercury-arc inverters, alternators and triode valve oscillators. More attention has justifiably been given to alternators and also to the design of valve oscillators, the theory of which is clearly presented. Many details of practical importance are explained. Most importance is attached to the use of h.f. generators for induction heating applications. The theory of induction heating is presented and many practical applications such as melting, hardening, brazing, annealing, sintering, etc., are described. The basic principles of dielectric heating are given, a few applications are briefly described, and some of the special circuit problems are explained. Much useful information is given about auxiliary equipment and h.f. measurements. Some worked examples and a description of some of the problems which arise from the operation of h.f. heating equipment add to the practical value of the book.

The bibliography is extensive and many illustrations of generators and equipment used for various processes are included.

R. SMITH

**The Principles of Rheological Measurement.** Report of a Conference arranged by the British Rheologists Club in 1946. (Edinburgh: Thomas Nelson and Sons, Ltd.) Pp. x + 214. Price 30s. net.

This book discusses the principles underlying the rheological properties of a wide range of materials and some new methods are described such as the pendulum viscometer and rheogoniometer. A useful analysis of hardness testing is given, although one feels that considerable progress has been made in this field since the Conference was held. The geometry of rheological phenomena is described in too general terms and it is a pity that the Table which illustrates some of the practical conclusions is not reproduced. A chapter is devoted to the anisotropy of wood and another dealing with the mechanical testing of solids is of exceptional interest. The large number of references, particularly those on pastes and doughs, should provide a valuable source of information.

W. LETHERSICH

**Atmospheric Turbulence.** By Prof. O. G. SUTTON, F.R.S. (London: Methuen and Co., Ltd.) Pp. viii + 107. Price 6s. net.

This is the first book entirely devoted to atmospheric turbulence to be published in this country and it is a very good one. Professor Sutton has treated this important aspect of dynamical meteorology from the standpoint of the mathematical physicist and his presentation of this difficult subject is masterly. For its size the book is a remarkably comprehensive survey of all the important investigations and theories of turbulence in the lower layers of the atmosphere.

A very good feature of the book is that it points out clearly the deficiencies of our knowledge of atmospheric turbulence and so indicates the lines along which future research can most profitably be directed.

F. J. SCRASE

**Radioactive Measurements with Nuclear Emulsions.** By H. YAGODA, B.Sc., Ph.D. (New York: John Wiley and Sons, Inc.; London: Chapman and Hall, Ltd.) Pp. ix + 356. Price 40s. net.

Yagoda's book deals with the use of photographic plates, particularly nuclear track plates in a variety of applications of radioactivity in metallurgy, mineralogy, petrography, biology, radiochemistry and nuclear physics. The author offers a wealth of useful and detailed information on subjects which are widely scattered in literature and his book can be regarded as a welcome contribution to the subject. Workers in the field of autoradiography will find detailed treatment of the following subjects: Preparations of specimens for

autoradiography, techniques of exposure, considerations of resolution, quantitative evaluation of autoradiographs, pseudo-photographic effects, properties of photographic emulsions in regard to sensitivity to various types of particles, to fading, loading and eradication of latent image.

The bulk of the contents comprises the description of the techniques used for the recording of alpha-particle tracks and patterns produced by natural radioactive sources. One cannot expect the author to be expert in every field in which the autoradiographic technique is applied, but it will be found rather disappointing by many readers that the biological applications, on which so much attention is at present focused in laboratories all over the world, are covered by only about 7% of the total pages. The recording of electron tracks could only be touched, through no fault of the author, since this important progress was made while the manuscript was being completed. The book is well produced and contains an excellent bibliography.

R. H. HERZ

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Special articles and Original Contributions accepted for publication

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## SPECIAL ARTICLE

## Environmental Warmth and Human Comfort

By T. BEDFORD, D.Sc., Ph.D., Medical Research Council, Environmental Hygiene Research Unit,  
London School of Hygiene and Tropical Medicine, Gower Street, London, W.C.1

The measurement of body heat production is discussed, and sample rates of heat production for various forms of activity are quoted. The physiological mechanisms which regulate the rate of heat loss are described. Various scales of warmth which take into account other environmental factors besides air temperature are described and their validity discussed in the light of the results of an extensive study of the thermal comfort of factory workers. The "comfort zone" is defined in terms of these scales of warmth. Reference is made to temperature gradients and to draughts as causes of discomfort. The factors which produce feelings of freshness or of stuffiness are discussed. Attention is drawn to the invigorating effect of variable air movement and of the combination of warm walls and cool air. The requirements for a pleasant and invigorating indoor environment are summarized.

## BODY HEAT PRODUCTION AND HEAT LOSS

The vital processes of the human body are accompanied by considerable energy exchanges. A man doing moderate work has total energy requirements amounting to roughly 3000 kg.cal., or about 12,000 B.Th.U./day. Much of this energy is evolved as heat and serves to maintain the body temperature. Even when the body is completely at rest and in warm surroundings, the heat production does not fall below a certain minimum level—the basal metabolism—and when work is being done the heat production rises considerably. The gross mechanical efficiency achieved varies with the type of work. For many tasks it is of the order of 20%, the remaining 80% or so of the energy expended appearing as heat. Thus it is that in cold surroundings we can keep warm by taking exercise.

One of the physiological requirements for health is the maintenance of a practically constant body temperature. In man, the average mouth temperature is about 98.4° F. and the average temperature of the deeper tissues is about 99° F. In health, these temperatures vary only slightly. Clearly, in order that the body temperature may remain constant it is necessary that the rate of heat production within the body and the rate of heat loss from it shall be equal, and elaborate provision is made to secure this balance. Physiological mechanisms regulate the heat production within the body and the rate of heat loss from its surface, so that over a limited range of ambient temperature the body temperature remains nearly constant. At environmental temperatures below this range the rate of heat loss must be limited by the use of clothing or the heat production increased by taking exercise.

A person of average size sitting at rest produces heat at the rate of about 350 B.Th.U./hr., but even slight activity raises the energy exchange, and for sedentary persons 400 B.Th.U./hr. is a reasonable figure. For some other grades of work typical figures of heat production are:

Type of work	Heat production, B.Th.U./hr.
Light work, standing	550
Light work, moving about	600–700
Walking at 3 m.p.h.	1050
Walking at 4 m.p.h.	1400
Walking at 5 m.p.h.	2300

The human body loses heat by radiation, convection and evaporation. As with an inanimate object, the rate of heat loss by these paths depends on the temperature, humidity and speed of movement of the air and on the radiation from the surroundings. It is also affected by the operation of the physiological controls.

*Regulation of Heat Loss.*

(i) *Skin Temperature.*—One of the heat-regulating mechanisms is that which varies the supply of blood to the skin. The reaction of the cutaneous circulation to thermal stimuli is complex but, in general, the blood vessels of the skin constrict with cold and dilate with heat. Thus in cold surroundings the blood supply to the skin is diminished and a lowered temperature of the skin surface results. Warmth provokes flushing of the skin with blood and the surface temperature rises. These variations of the temperature of the skin surface modify the effects of the environment upon the rate of heat loss by convection and radiation. Except in warm conditions the skin temperature of the extremities is usually distinctly lower than that of the head and trunk. Various observers have studied the conductance of the human skin. It appears that the conductance reaches a minimum when the average skin temperature is about 8° F. below the temperature of the body tissues as indicated by the rectal temperature. With further cooling of the skin the conductance is unchanged. As the gradient is reduced below 8° F. by exposure to warm conditions the conductance increases rapidly.

(ii) *Evaporation.*—The body also loses heat by the evaporation of water from the skin and lungs. The expired air is saturated with water vapour and the latent

heat of vaporization, together with the heat required to warm the air, is taken from the respiratory surface. Water is lost from the skin by sweating at high temperatures and by insensible perspiration in cooler surroundings.

Sweating is due to the activity of the sweat glands in the skin. It may be regarded as an emergency mechanism which operates when the heat loss by other paths is insufficient to dissipate the heat produced by the body. The term "insensible perspiration" is used to denote the loss of water from the skin when no sweating occurs, together with that from the lungs. The insensible loss from the skin occurs even in a cool environment when the skin is apparently dry, but it increases in warmer surroundings which are yet not warm enough to stimulate the sweat glands to activity. It has been shown to be an osmotic process.

#### *Measurement of Body Heat Production.*

Direct measurements of the total heat loss from the body are made by placing the subject in some form of calorimeter. Heat production is measured indirectly by measuring the oxygen consumption and the carbon-dioxide production and then computing the heat production from these values. Crawford used a calorimeter to measure the heat production of a guinea pig as early as 1777 and a few years later Lavoisier repeated the animal experiments.

In the modern calorimeter heat production and heat loss are measured simultaneously. If the mean temperature of the body remained constant the heat production would balance the heat loss, even in experiments of short duration. Actually such equilibrium is probably rarely achieved, yet the temperature-regulating mechanism keeps the changes of body temperature within narrow limits, so that in prolonged experiments in the calorimeter it is found that measurements of heat loss and of heat production agree very closely. Although the direct measurement of the total heat loss by calorimetry is simple in theory, it is difficult in practice and thousands of measurements are needed during the course of a 3 hr. experiment. Yet, in spite of these difficulties, DuBois and his colleagues have carried on valuable researches over many years with the aid of the Sage Calorimeter at the New York Hospital. For most purposes, however, body heat production is usually measured by the indirect method and the heat loss is calculated by allowing for storage when the body temperature is not constant.

#### *Apportionment of Body Heat Losses.*

For many years physiologists have interested themselves in the partitioning of body heat losses. The evaporative heat loss is readily calculated from measurements of weight loss so that, if the total heat loss is known, the sum of the radiation and convection losses is known. Various researches carried out during the past half-century or so have shown that at ordinary room temperatures the heat loss by evaporation from a resting person is between 20 and 30% of the total heat

loss. The apportionment of the remaining 70 or 80% between radiation and convection has yielded conflicting estimates.

#### SCALES OF WARMTH

It is the common practice to specify the thermal environment in terms of the air temperature only, or of the temperature and humidity. From time to time, however, attempts have been made to devise scales of warmth which would take into account other environmental factors and a brief account of some of these is desirable.

#### *Kata Thermometer Cooling Power.*

The kata thermometer was introduced by Sir Leonard Hill (1, 2) as an instrument for assessing the physiological effects of the environment. It is an alcohol thermometer with a bulb about 4 cm. long and 2 cm. in diameter. The thermometer is heated and the rate of cooling from 100° F. is ascertained. This rate of heat loss is referred to as the "dry kata thermometer cooling power." The smallness of the instrument makes it very sensitive to the effects of air movement and suitable for measuring air currents, but that sensitivity renders it unreliable as an index of the effects of warmth on the human body.

#### *Effective Temperature.*

A scale of warmth which is widely used by heating and ventilating engineers, especially in North America, is effective temperature. From an alinement chart one can read off the effective temperature represented by any combination of air temperature, humidity and speed of air movement. The effective temperature of an environment connotes that temperature of still air, saturated with water vapour, in which an equivalent sensation of warmth was experienced in tests carried out by Yagloglou and his colleagues (3, 4, 5) in the Pittsburgh laboratory of the American Society of Heating and Ventilating Engineers. A criticism of the effective temperature scale is that it makes no specific allowance for radiation. The scale is applicable to conditions in which the air and the surroundings are at the same temperature, but, if the temperature of the radiating surfaces differs sensibly from that of the air, some correction for radiation is clearly necessary.

#### *Equivalent Temperature.*

The need for an instrument which, while taking into account radiant heat, would correspond approximately to the human body in its sensitivity to air currents, led Dufton (6) to construct a blackened copper cylinder, the eupatheostat, for use as a radiation thermostat. In the form of the eupatheoscope this instrument was then developed as a method of assessing the thermal environment (7, 8). The eupatheoscope is a heated cylinder 22 in. high and 7½ in. in diameter. In the original form of the instrument a bimetallic strip thermostat inside the cylinder was maintained at 78° F. In still air with the surroundings at 65° F., the surface temperature of



the eupatheoscope was  $75^{\circ}\text{F}$ ., which is about the average surface temperature of the clothed human body under such conditions. In its present form (Mark II), the instrument has a surface temperature almost identical with that of the clothed human body over a considerable range of environmental temperatures (9).

The readings of the eupatheoscope are expressed on a scale of *equivalent temperature*. The equivalent temperature, as indicated by the Mark II instrument, can be defined as: That temperature of a uniform enclosure in which, in still air, a black body of sufficient size would lose heat at the same rate as in the environment, the surface temperature of the body being one-third of the way between the temperature of the enclosure and  $100^{\circ}\text{F}$ .

### *The Globe Thermometer.*

Vernon (10) introduced his globe thermometer as a means of indicating the combined effects of radiation and convection as they affect the human body. The globe thermometer consists of a hollow 6 in. copper sphere, coated with matt black paint, and containing an ordinary thermometer with its bulb at the centre of the sphere. In the calm conditions which usually prevail in occupied rooms the globe thermometer is a simple but very useful index of comfort, for it takes into account both air temperature and radiant heat. The instrument is inadequate, however, when used over a range of air speeds. Thus, if the surroundings are cooler than the air the globe thermometer will be below air temperature. If, then, the radiation and air temperature are held constant and the air speed increased, the temperature of the globe will approach more nearly to the air temperature, i.e. it will rise, although with the higher air speed the cooling effect on the human body will be greater.

### THERMAL COMFORT OF INDUSTRIAL WORKERS

Early in the nineteenth century, Tredgold (11) wrote: "... the art of applying heat has been studied with attention, and illustrated with talent; while it has been practised by men of no ordinary skill, yet still there appears to be a field sufficiently open for new and useful researches; for it seems to be possible to combine an equal degree of safety, cleanliness, and comfort with more healthiness and economy."

Research is still necessary, as new methods of heating are introduced or old ones modified. Some years ago the growing use of novel methods of radiant heating made it necessary that more attention should be paid to the study of human comfort in relation to the warmth of the surroundings and I undertook a study of the comfort of industrial workers (12).

The study was carried out in twelve factories in a variety of industries. The buildings and the methods by which they were warmed and ventilated were as varied as the manufacturing processes carried on in them. The observations were confined to the winter months when artificial heating was in use. At each observation

position instrumental measurements of the thermal environment were made, while the workers in the immediate neighbourhood were carefully questioned as to their thermal comfort. Measurements were then made of the skin temperatures of a hand, a foot, and the forehead of each worker and of the mean surface temperature of his or her clothed body. In all, over 3000 sets of observations were made, 94% on women and girls and the remainder on men and boys. Nine-tenths of the subjects were seated at their work but had to walk from time to time to fetch materials. All were engaged on very light work.

The workers' responses to the questioning were classified on a seven-point scale:

<i>Sensation of warmth</i>	<i>Numerical scale</i>
Much too warm	1
Too warm	2
Comfortably warm	3
Comfortable	4
Comfortably cool	5
Too cool	6
Much too cool	7

"Comfortably warm" meant that the subject was not uncomfortable but would have preferred a somewhat cooler environment and, similarly, "comfortably cool" indicates a preference for a little more warmth. For the purpose of statistical treatment numerical values were assigned, as indicated above.

The temperature, humidity and speed of movement of the air, and the radiation from the surroundings were measured at a height of 4 ft. from the floor, i.e. at about the head level of a person seated at a work-bench, and further measurements of temperature and air speed were made 6 in. above the floor. Air temperatures were measured by means of mercury-in-glass thermometers, the bulbs of which were screened from radiation by aluminium foil, and whirling hygrometers were used for humidity measurements. For the measurement of the low air speeds which usually prevail indoors, kata thermometers with silvered bulbs were used. A Moll thermopile was used for the radiation measurements. The thermopile was pointed in turn to every part of the sphere surrounding the observation point and the mean intensity of radiation thus calculated. The radiation intensity was expressed as mean radiant temperature. In addition, the warmth of the environment was expressed in terms of dry kata cooling power, equivalent temperature and effective temperature and globe thermometer readings were taken.

The ranges of conditions encountered, in terms of the basic measurements at 4 ft. from the floor, were:

<i>Thermal measurement</i>	<i>Range</i>	<i>Mean</i>	<i>Standard deviation</i>
Dry bulb temperature, $^{\circ}\text{F}$ .	54-76	64.4	3.67
Mean radiant temperature, $^{\circ}\text{F}$ .	54-80	67.1	4.62
Partial pressure of water vapour, mm. mercury	5-14	7.2	1.11
Air speed, ft./min.	10-100	31	14.6

The range of air temperature shown is very similar to that of the mean radiant temperature, but at individual observation positions there were considerable differences. Sometimes the mean radiant temperature was 4° to 5° F. below air temperature, whilst in others it was as much as 15° F. above it.

We all know that there is wide diversity of opinion as to what constitutes comfortable warmth and this was evident in this investigation. At an air temperature of 64° to 66° F. most people were quite comfortable, and 92% of the persons were "comfortable," "comfortably warm," or "comfortably cool," i.e. only 8% were uncomfortable, yet of these some were too warm and some too cool.

#### *Correlation of Comfort Votes with Scales of Warmth.*

One object of the study was to examine the value of the various scales of warmth which have been described. The correlation between comfort votes (using the numerical marks mentioned earlier) and the degree of warmth as measured by the different indices is shown below.

<i>Comfort votes correlated with</i>	<i>Correlation coefficient (r) and standard error</i>
Equivalent temperature	- 0.52 ± 0.01
Globe thermometer reading	- 0.51 ± 0.01
Effective temperature	- 0.48 ± 0.01
Air temperature (dry bulb)	- 0.48 ± 0.01
Mean radiant temperature	- 0.47 ± 0.01
Dry kata cooling power	+ 0.43 ± 0.01
Absolute humidity	- 0.12 ± 0.01

These correlations are based on 2571 sets of observations. Data obtained in one large factory were excluded from these calculations because of certain special local conditions and were submitted to separate examination. The first four correlation coefficients did not differ significantly from each other, indeed there was no difference of practical significance between the first five. Yet it is of some interest that equivalent temperature, which makes due allowance for radiation, and air movement, as well as air temperature, gave the highest correlation, whilst, since there was no wide range of air movement, the globe thermometer was a close runner-up. As a measure of warmth the kata thermometer was inferior to the simple dry-bulb thermometer. Atmospheric humidity was significantly correlated with warmth sensations, increased humidity being associated with feelings of greater warmth, but the association was not a close one. Although atmospheric humidity is of outstanding importance when the temperature is high, at ordinary room temperatures it exerts little effect on our feelings of warmth.

#### *The Comfort Zone.*

In view of the diversity in the comfort votes recorded at any room temperature it is obviously impossible to fix any particular condition of warmth that will be acceptable to everybody, but one can ascertain what conditions are likely to be most generally acceptable.

This information can be obtained from regression equations relating comfort votes to the various measures of warmth. The most comfortable conditions can be taken as those which according to the regression equations yield an average vote of "comfortable," equalling 4 on the arbitrary numerical scale. The most comfortable conditions for the persons doing very light work, with which this inquiry was concerned, were:

<i>Scale of warmth</i>	<i>Most comfortable degree of warmth (°F.)</i>
Equivalent temperature	62.3
Globe thermometer temperature	65.1
Effective temperature	60.8
Air temperature (dry bulb)	64.7

Unless the temperature of a room is automatically controlled fluctuations will occur. The temperature of a workroom may vary considerably during the course of a day. It is important, therefore, that we should know between what limits the temperature should be allowed to vary; we should define the comfort zone. Various workers have described comfort zones determined by different standards. Most frequently limits have been defined within which not less than 50% of the persons questioned felt comfortable. Such limits seem to be too wide for, as they are approached, a substantial proportion of persons will experience actual discomfort.

On the basis of my data I recommended that the comfort zone should be taken as that in which not less than 70% of the subjects were "comfortable." In that range at least 86% of the votes ranged from "comfortably cool" to "comfortably warm," so that at no temperature within this zone did more than 14% experience discomfort. The zone is:

<i>Scale of warmth</i>	<i>Temperatures (°F.) between which not less than 70% of persons were "comfortable"</i>
Equivalent temperature	58-66
Globe thermometer temperature	62-68
Effective temperature	57-63
Air temperature (dry bulb)	60-68

These temperatures are distinctly lower than those found desirable for Americans, partly because the indoor clothing worn in Great Britain is usually warmer than that worn by Americans and partly, probably, because in Great Britain we are acclimatized to lower indoor temperatures.

#### *Effects of the separate Thermal Factors.*

In view of the differences between the scales of warmth which have been described, it was of considerable interest to ascertain the effects of the separate thermal factors on sensations of warmth. It was clear that the influence of air movement must vary with variations in the surface temperature of the body. Observations had shown that the difference between mean body surface temperature and air temperature could be expressed in terms of  $(100 - t_a)$ , where  $t_a$  is the air temperature, and there was evidence that the cooling effect of wind



varied as the square root of the air speed. Hence in deriving an equation for these observations a term in  $\sqrt{v}(100 - t_a)$  was used.

When in turn, air temperature ( $t_a$ ), mean radiant temperature ( $t_w$ ), vapour pressure ( $f$ ), and the convection function  $\sqrt{v}(100 - t_a)$ , were correlated with comfort votes, the remaining factors being held constant, each correlation coefficient was statistically significant. From these coefficients an equation expressing comfort votes in terms of the four thermal factors was derived:

$$S = 11.16 - 0.0556t_a - 0.0538t_w - 0.0372f + 0.00144\sqrt{v}(100 - t_a)$$

where  $S$  is the comfort vote on the numerical scale described earlier,  $t_a$  is the air temperature, in °F.,  $t_w$  is the mean radiant temperature, in °F.,  $f$  is the partial pressure of water vapour in the air, in mm. of mercury, and  $v$  is the air speed in ft. per min.

In still air the effect on warmth sensations of a given change in the mean radiant temperature is practically the same as that of a similar change in the air temperature. As the air speed increases, the effect of the temperature of the surrounding surfaces relative to that of the air decreases. With air moving at 20 ft./min., a rise of 8.7° F. in the air temperature will compensate for a fall of 10° F. in the mean radiant temperature, whereas with an air speed of 100 ft./min. the compensatory rise in the air temperature is 7.7° F.

With constant vapour pressure, an enclosure with air and walls at 65° F. and with the air moving at 10 ft./min. feels as warm as an enclosure at 67.8° F. in which the air speed is 100 ft./min.

#### *Calculation of Equivalent Temperature.*

Since, at normal room temperatures, a moderate change in atmospheric humidity has only a small effect on subjective sensations of warmth, no serious error is incurred by ignoring humidity in assessing environmental warmth at such temperatures. Equivalent temperature, which takes no account of humidity, but allows for the other thermal factors, is a valuable index of warmth under such conditions. From the data obtained in this inquiry an equation was derived from which equivalent temperature can be estimated with reasonable accuracy when the component thermal factors are known. It is:

Equivalent temperature

$$= 0.522t_a + 0.478t_w - 0.01474\sqrt{v}(100 - t_a)$$

Although a scale of warmth such as equivalent temperature is of much value for the comparison of environments it is highly desirable that, when this or any other scale of warmth which combines the effects of two or more thermal variables is used, the individual thermal measurements should be known. Then, if the observed conditions are unsuitable, examination of the individual measurements should reveal the direction in which a remedy should be sought. To calculate equivalent temperature from this equation the mean radiant temperature must be known. If no suitable radiometer is available,

this can be estimated with satisfactory accuracy by means of the globe thermometer (13).

#### *Some Sources of Discomfort.*

(i) *Temperature Gradients.*—Methods of warming and ventilating rooms which give distinctly greater warmth about the head than near the floor are undesirable, for such a condition tends to produce a stuffy feeling in the head whilst the feet are cold. This has been appreciated for over a hundred years yet, even in recent times, systems of heating which set up steep temperature gradients have been widely used, both in industrial buildings and in dwellings.

(ii) *Draughts.*—The speed at which an air current becomes noticeable depends on its temperature. At temperatures of 60° to 65° F. currents impinging on the cheek with speeds below about 40 ft./min. are not perceptible; at 54° F. a current of 30 ft./min. can just be felt, whilst at 86° F. currents of less than 120 ft./min. are unnoticed (14).

In my investigation the maximum air speed was about 100 ft./min. and mostly the speed was below 50 ft./min. Few complaints were made of draughts, but some persons complained even when the air speed was below 20 ft./min. The people who complained had skin temperatures rather below the average and, probably partly on that account, they had a distinct preference for a warmer environment than sufficed for their fellows.

#### FRESHNESS AND STUFFINESS

So far we have discussed environmental conditions in relation to sensations of general bodily warmth. It remains to consider those conditions which lead to feelings of freshness or of stuffiness. Of two rooms with the same general level of warmth, say, with the same equivalent temperature, one may seem fresh and the other stuffy. These feelings of freshness are mainly conditioned by the thermal environment, and they are thus to some extent correlated with sensations of warmth. A cool room tends to feel fresh and an overheated one stuffy. Feelings of general warmth depend on the summated effects of the environment on the surface of the whole body. Impressions of freshness and of stuffiness depend on the stimulation or lack of stimulation of areas of skin. Pressure changes due to sudden changes in the strength or direction of air movement may be enough to stimulate the tactile nerve-endings in the skin, and thus to arouse touch sensations. This is a common experience out of doors, but indoors the changes in air speed are commonly not large enough to arouse recognizable touch sensations. Yet changes in air speed such as are commonly encountered in well-ventilated rooms are sufficient to stimulate the thermal receptors in the skin. Likewise, variability of the other thermal factors may also contribute to such stimulation.

Vernon, Bedford and Warner (15) studied the effects of air movement and temperature on impressions of freshness, and the data were re-examined by Bedford and

Warner (16). Freshness was correlated with air temperature, a cool room tending to feel fresher than a warm one. When temperature was held constant, there was a clear effect of mean air speed, higher air speeds being associated with feelings of greater freshness. When air temperature and mean air speed were both held constant, increased variability in the air speed was associated with enhanced freshness.

When mean air temperature and air speed were kept constant the partial correlation coefficient for freshness and temperature variations was insignificant. In 73% of the observations the mean variation of temperature, in measurements made at  $2\frac{1}{2}$  sec. intervals, was only  $0.4^{\circ}$  F., and these slight variations had no apparent influence on sensation. There was a significant relationship between relative humidity and freshness at constant air speed and temperature; increased humidity tended to cause feelings of stuffiness. It appeared from these observations that a change in humidity which is insignificant from the standpoint of general warmth sensations may have a distinct effect on one's feelings of freshness or of stuffiness.

Hardy and Oppel (17) found that the smallest intensity of radiation which could be perceived when the skin was irradiated for 3 sec. was  $0.00015$  cal./sq. cm./sec. Such a change in radiation intensity would be caused if the temperature of the surfaces radiating on the skin were suddenly raised by  $2^{\circ}$  F., the temperature, speed and humidity of the air remaining unchanged. Changes in these atmospheric conditions which would bring about corresponding shifts in the convection exchange or in the evaporative cooling would also cause sensations.

It has long been held that, from the standpoint of comfort, the walls of a room should be warmer than the air rather than cooler. C. G. Warner and I (16) held that view. Although we had no statistically valid evidence on the point, we had experienced feelings of stuffiness when we were unable to find any satisfactory explanation other than the fact that the mean radiant temperature was  $6^{\circ}$  F. or more below the air temperature.

Recent work by Munro and Chrenko (18) has provided reliable evidence that with the same equivalent temperature warm walls and cool air give an environment which feels fresher than one in which the walls are cooler than, or at the same temperature as, the air. This is thought to be due mainly to the fact that the partial pressure of water vapour in the air was lower in the warm-wall experiments than in the others. It is considered by these workers that freshness impressions are related to transient fluctuations in the rate of heat loss from the head.

We can sum up the requirements for a pleasant and invigorating indoor environment as:

- (i) A room should be as cool as is compatible with comfort.

- (ii) There should be adequate air movement, but no local draughts. At the temperatures commonly maintained in British buildings during cold weather air speeds much below 20 ft./min. tend to cause feelings of stuffiness, whilst distinctly higher speeds are desirable in warm weather.
- (iii) The air movement should be variable, rather than uniform and monotonous.
- (iv) The relative humidity of the air should not exceed 70%, and should preferably be well below that figure.
- (v) The mean temperature of the solid surroundings should preferably be above the air temperature.
- (vi) The air at head level should not be distinctly warmer than that near the floor and the heads of the occupants should not be exposed to excessive radiant heat.

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## ORIGINAL CONTRIBUTIONS

## Improvements in Photoelastic Technique Obtained by the Use of a Photometric Method

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A photoelectric photometer has been adapted for use in photoelasticity. The sensitivity of the instrument is such that a photoelastic stress pattern can be measured to within  $\pm 1/500$  fringe. The resultant improvements in technique are: (1) the use of materials having less susceptibility to edge effects than those generally used, but with too low a stress-optical coefficient for examination by normal methods; (2) the use of lower stresses which should be of particular advantage in three-dimensional stress analysis by the frozen stress method, where lower stresses would reduce the strains which are often excessive in this method; and (3) greater ease in tracing isoclinics because the photometer enables a distinction to be made between isoclinics and isochromatics. The apparatus is described, examples of its use are given and factors limiting its accuracy are discussed.

In the course of their work at the National Physical Laboratory the authors have from time to time felt the desire for greater accuracy in determining the stress from the isochromatic pattern. In general, the need arises from one of two causes, either from the use of a low stress or of a model material having a low stress-optical coefficient.

Methods of compensation are used in which a measured double refraction effect is superposed at the point where the stress is required, so as to balance the retardation due to stress. Examples are the Babinet compensator using two quartz wedges and the Coker type which consists of a tensile specimen made of photoelastic material. In the Tardy method the analyser is rotated so as to move the fringes across the picture. All these methods rely on visual judgment of the best degree of extinction for adjustment of the amount of compensation. It is believed that some experimenters claim a high degree of accuracy for this operation but the authors, not being satisfied with their own performance, thought that some kind of photometer might be used for detecting the condition of minimum light.

## PHOTOMETER METHOD

The design of the photometer used for the work was based on an instrument developed in the Photometry Section of the Light Division of the Laboratory. The first trials were so encouraging that it was decided to have a similar instrument made specially for photoelastic work.

Fig. 1 shows the screen on to which the photoelastic pattern is projected from a normal optical bench provided with a circular polariscope. The photometer is mounted behind the square aluminium plate which has a central hole into which circular plates having apertures of various diameters can be inserted. The whole board can be

moved in its own plane horizontally and vertically so that any part of the field can be brought over the aperture, the size of which determines the fineness with which the pattern can be examined. As the magnification

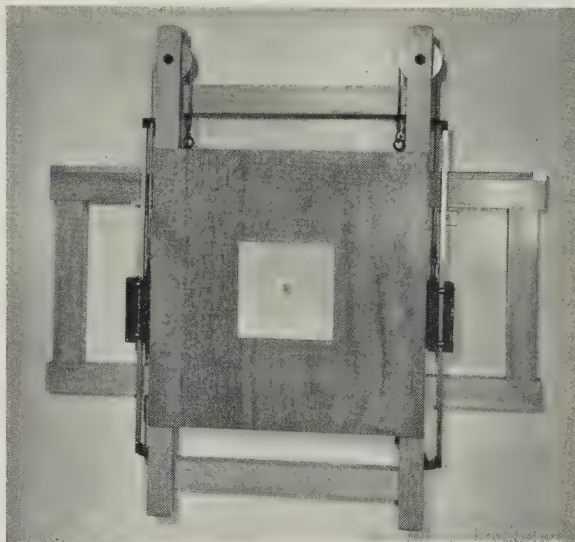


Fig. 1. Adjustable screen with photometer mounted centrally

from model to screen is about 9:1, a  $\frac{1}{8}$  in diameter aperture corresponds with a diameter of about 0.014 in on the model.

Fig. 2 shows the way in which the photometer can be rotated for access to the components. An airtight cover has been removed from the container housing a photocell and electrometer valve which are kept dry with silica gel to reduce electrical leakage. Fig. 3 is a diagram of connections. The photocell, chosen for its sensitivity to green light, allows current to pass through the 5 000 M $\Omega$  resistance when its cathode is illuminated

and the resulting potential, fed to the electrometer valve, causes the galvanometer to deflect. Thus a minimum galvanometer deflexion indicates minimum light. In

## APPLICATIONS

(a) *Use of Low Sensitivity Materials.*—One application of the method is to the use of low sensitivity materials which may have advantages such as freedom from edge effects to counter-balance their poor response to loading. One example is glass, the material on which pioneer photo-elastic experiments were made. Residual stresses can be made negligible by annealing and machining stresses could probably be removed by further annealing, but hand finishing does not appear to leave any edge effects. As far as the authors are aware, there is no time-edge effect.

To determine whether satisfactory stress measurements could be made with glass, a circular disk of

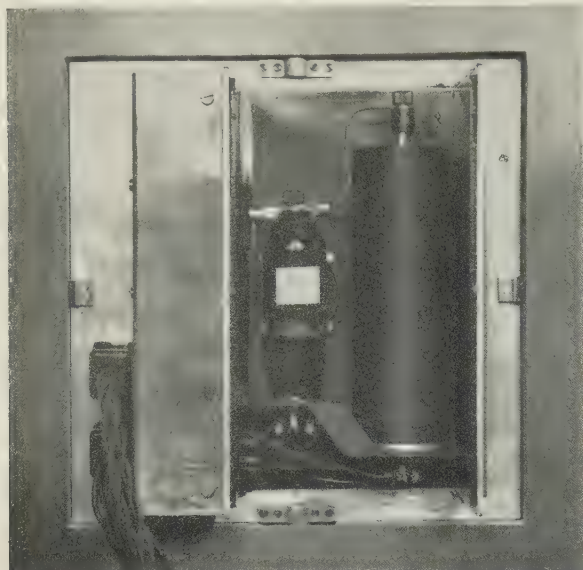


Fig. 2. Rear view of photometer with cover removed

practice the galvanometer, which may be of the portable reflecting type, is adjusted to zero deflexion with no light by altering the  $500\ \Omega$  potentiometer, the  $1\ \text{M}\Omega$  potentiometer being used to balance any stray light under working conditions. As an indication of the sensitivity, it is estimated that with a  $\frac{1}{8}$  in aperture the light from

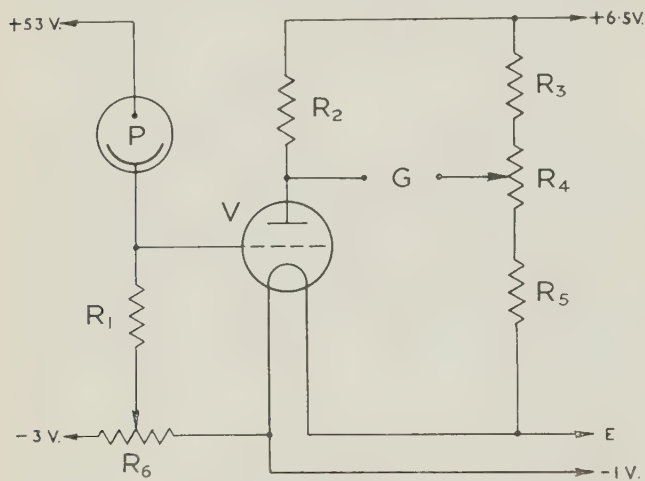


Fig. 3. Circuit diagram of photoelectric photometer

$R_1$ ,  $5\ 000\ \text{M}\Omega$ ;  $R_2$ ,  $10\ 000\ \Omega$ ;  $R_3$ ,  $1\ 500\ \Omega$ ;  $R_4$ ,  $500\ \Omega$ ;  $R_5$ ,  $2\ 200\ \Omega$ ;  $R_6$ ,  $1\ \text{M}\Omega$ ; P photocell, VA26T; V, type ET1.

a candle could be detected at 100 yd. The photometer can be used to detect the middle of a fringe by moving the screen bodily until the galvanometer gives its minimum reading. Alternatively, with the photometer fixed, the null position of the galvanometer spot gives a clear indication of the correct setting of the compensator.

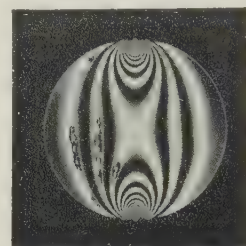


Fig. 4. Stress pattern in circular Bakelite disk under diametral compression. Diameter of disk, 1.07 in; thickness of disk, 0.258 in; load, 95 lb approx.

crown glass,  $\frac{1}{4}$  in thick, was prepared and examined with mercury green light. Fig. 4 shows the familiar stress pattern in a  $\frac{1}{4}$  in thick disk of Bakelite BT 61 893 under diametral compression and Fig. 5 shows the glass disk loaded to rather less than half the stress—the principal stress difference at the centre was  $400\ \text{lb/in}^2$  corre-

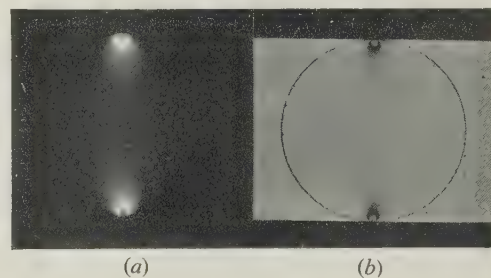


Fig. 5. Stress pattern in circular glass disk under diametral compression: (a) dark field; (b) light field. Diameter of disk, 1.08 in; thickness of disk, 0.272 in; load, 46 lb.

sponding with 0.09 fringe, so that the glass had a stress-optical coefficient of 2.6 Brewsters. The difference between the principal stresses in the plane of the disk was measured over a quadrant by the Tardy method (1). After the isoclinics had been determined by the second method described in section (c) below, the value of the isoclinic parameter could be estimated with sufficient accuracy at any point in the quadrant. When the polarizer had been turned from zero through the isoclinic angle and the quarter wave plates rotated through the same angle, the analyser was moved until the photo-



meter indicated minimum light. The difference between the analyser angle in this position and the zero value near of the disk, when divided by  $180^\circ$ , gives the fraction of a fringe at the point examined.

Fig. 6 shows a comparison of the calculated and experimental values. The curves give the ratio of

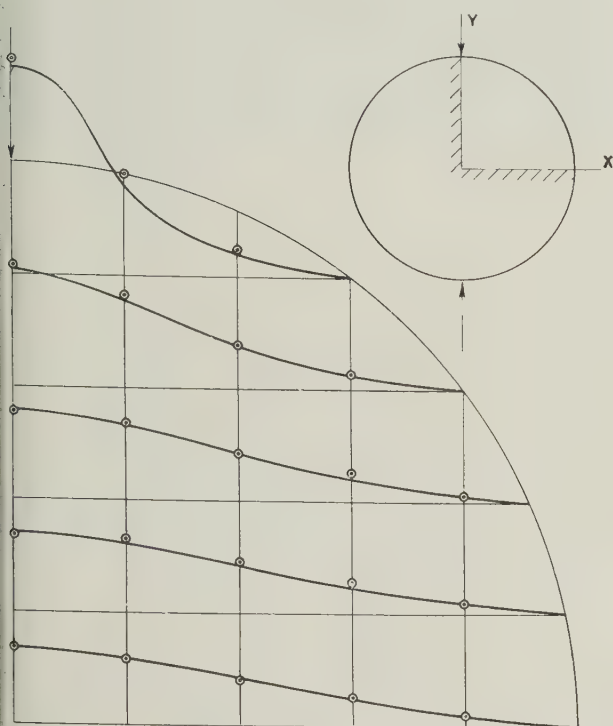


Fig. 6. Circular glass disk under diametral compression. Comparison of calculated and observed values of difference between principal stresses (the curves and points show respectively calculated and observed values along lines parallel to the X axis).

$-q$  along lines parallel with the X-axis to  $p - q$  at the centre, calculated from the expression:

$$p - q = -\frac{2Pd}{\pi t} \left( \frac{\frac{1}{4}d^2 - x^2 - y^2}{(\frac{1}{4}d^2 + x^2 + y^2)^2 - d^2y^2} \right)$$

where  $p - q$  = difference of principal stresses

$P$  = load applied

$d$  = diameter of disk

$t$  = thickness of disk

$x$  and  $y$  are co-ordinates of points in the disk,  $y$  being the axis of loading.

The points are the observed values and, considering that the stress gives under 1/10 fringe at the centre, the agreement is good. Some departure can be seen near the point of loading and this may be accounted for by the use of rather a large aperture ( $\frac{1}{4}$  in, corresponding with a circle on the disk having a diameter of about  $\frac{1}{40}$ ). The method gives the average value over the aperture and this will be greater than the stress at the

centre of the aperture where there is a high gradient towards the point of loading.

(b) *Use in Frozen Stress Work.*—The higher precision attainable with the photometer should be of value in frozen stress work. There are three possibilities: (1) application of smaller loads to sensitive materials such as Bakelite, thus reducing the strains which are considerable in the normal technique; (2) the use of thinner slices in regions where the boundary stresses are needed at an edge which is not perpendicular to the plane of slicing; (3) the possibility of using materials of low sensitivity which may not be so susceptible to "rind" or time-edge effects.

A case occurs in a recent investigation by W. A. P. Fisher of the stresses in a truncated Catalin cone loaded in compression, where it would be an advantage to use a thin slice. The higher stresses were in axial planes, but to complete the analysis it was necessary to find the

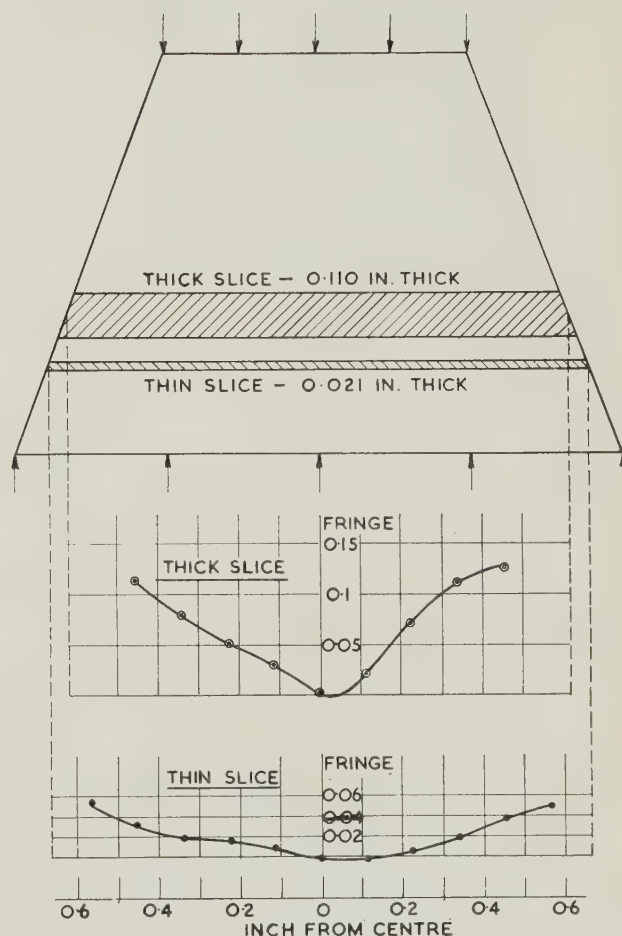


Fig. 7. Truncated cone in compression. Relative retardation in transverse slices.

difference between the radial and circumferential stresses by cutting a transverse section. Fig. 7 shows the dimensions of a truncated cone which was loaded with approximately uniform compression, heated at  $81^\circ\text{C}$  for 4 hr

and cooled under load. Transverse slices 0.11 and 0.021 in thick were cut and the fringe values were determined along a diameter with the photometer, using mercury-green light, the results being shown on the curves below. The actual values, particularly in the case of the thin slice, are very small, but it is significant that readings could be repeated to within  $\pm 1/500$  fringe. The original object, to determine accurately the boundary stress, is somewhat lost through the rind effect produced by the long heating of Catalin but this would not apply in a material free from this defect. Fig. 8 shows the stress pattern in the thicker (0.11 in) slice. With the thinner slice it was scarcely possible to detect visually any difference of tone.



Fig. 8. Stress pattern in transverse slice cut from truncated Catalin cone after compressive loading at  $81^{\circ}\text{C}$  (dimensions of cone as in Fig. 7, load 22.5 lb.)

(c) *Isoclinics*.—All those familiar with photoelastic work know the difficulty of tracing isoclinics. Drawing the curves by hand with the model under light load avoids the confusion of the isochromatics but is open to the objection that, in regions of low stress, residual stresses may distort the isoclinic direction to a considerable extent. Fortunately the photocell, being sensitive to a wide band of light frequencies, is able to discriminate between the coloured isochromatics and black isoclinics so that the mapping of the latter can be carried out under any convenient load.

Two methods are available, the first being to determine the isoclinic parameter at a given point by rotating the polarizer and analyser together till the isoclinic shadow falls centrally over the photocell aperture. In the second method the polarizer and analyser are set to a required parameter and the photometer is moved about so as to trace the course of the whole isoclinic. The moving photometer method gives the normal family of curves directly, but suffers from possible inaccuracy if the field is not uniformly illuminated.

Fig. 9 gives an example of the use of the second method to trace the  $20^{\circ}$  isoclinic in a simply supported beam, made in this case from Columbian resin CR39 and examined in white light. The reproduction, made on a

photographic plate without colour correction, shows some confusion between stress pattern and isoclinics but the dotted line indicates that the photometer can locate the centre of the dark bands with accuracy. The

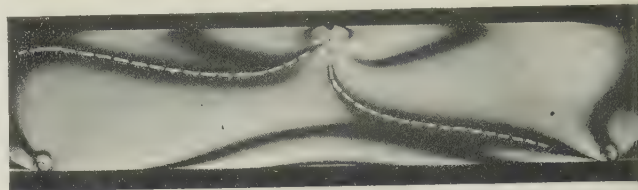


Fig. 9.  $20^{\circ}$  isoclinic in simply supported Columbian resin CR39 beam (centre line of isoclinic determined with photometer)

polarizer and analyser were adjusted to show the  $200^{\circ}$  isoclinic and the screen was moved till the galvanometer gave a minimum deflection. Co-ordinates were then measured from reference marks so that the isoclinic could be reconstructed to any convenient scale.

For the Tardy method of compensation, it is necessary to know the value of the isoclinic parameter at a certain point in the model and the first method is better, but in this case it is essential to be able to drive the polarizer and analyser together from a single spindle.

#### PARTICULAR APPLICATION

An example is given in Fig. 10 of a case where it is considered that the photometric method will be particularly useful. The problem is to find the value of the stress concentration at the base of a very shallow groove in a plate subjected to compression. The authors' experience in photoelasticity leads them to doubt whether edge effects in normal photoelastic materials would permit an accurate result. It is proposed therefore to do the experiment with glass where edge effects should not be troublesome, but where the low stress optical coefficient necessitates a refined method of measurement at the boundary of the groove.

#### LIMITS OF ACCURACY

It has been stated that the stress pattern can be measured to within  $\pm 1/500$  fringe. This is based on the experience that, when using the Tardy method, analyser readings can be repeated to within  $\pm \frac{1}{4}^{\circ}$ . For a comparison with photometer results a number of readings were taken on one spot on the Bakelite disk loaded in compression and examined in mercury-green light, judging by eye the greatest degree of extinction. Readings were then taken with the photometer on the same place in the disk using an  $\frac{1}{8}$  in diameter ( $d/80$ ) aperture. Twelve readings were recorded by each of three observers and the averages of the results are taken in groups of three as being representative of a routine test.



*Visual determination by the Tardy method of principal stress difference at an arbitrary point in Bakelite disk loaded in compression*

Average values of analyser angle (4 groups of 3 observations)

Observer	1st group	2nd group	3rd group	4th group	Average of 12 observations
A	327.8	327.3	330.2	330.2	328.9
B	327.2	327.2	330.5	325.2	327.5
C	330.3	330.2	333.3	333.2	331.8

The rather wide scatter in the visual results may be explained by the fact that the point chosen for examination was not on one of the axes of symmetry. This

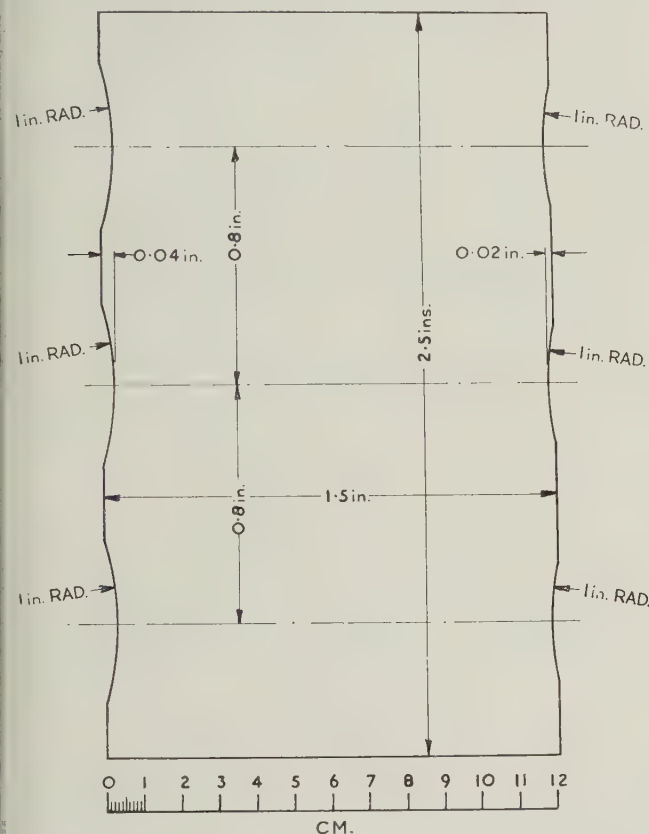


Fig. 10. Dimensions of plane model for determination of stress concentrations caused by shallow grooves

increased the difficulty of assessing the best degree of extinction and resulted in a range of over  $4^\circ$  in the grand averages for the three observers. For comparison a number of repeated measurements were made by two observers with the photometer and these gave figures not differing by more than  $\frac{1}{4}^\circ$ , the average value being  $328.6$ .

Such precision as is shown in the photometer readings demands a high degree of stability; in fact, the degree of accuracy of the photometer is determined by the steadiness of the galvanometer spot. The question of electrical stability is discussed in the Appendix, but a very stable light is needed in view of the minute changes of light which have to be detected. This need could be avoided by using a balanced system with two photocells, one recording directly from the source and the other

from the stress pattern. In the authors' equipment a single cell was used and it was found that to realize the accuracy mentioned above it was necessary for the light to be constant to within 2%, which required a voltage constancy at the lamp terminals within less than 1 part in 200. This was achieved by the use of a stabilized transformer which was found to be essential in all the measurements made.

Mindlin (3) gives a general expression for the intensity of light transmitted by a polariscope and, for the case of a perfect circular polariscope, this reduces to:

$$I = I_0 \sin^2 \left( \theta - \frac{\delta}{2} \right)$$

where  $I$  is the intensity of light transmitted with analyser angle  $\theta$ ,  $I_0$  is the maximum intensity of light transmitted,  $\theta$  is the analyser angle measured from the extinction position with no specimen,  $\delta$  is the phase retardation introduced by the specimen. With a specimen in position, no light passes when  $\theta = \delta/2$  and if the analyser is rotated, the light passing varies as the square of the sine of the angle turned through.

In a normal imperfect polariscope some light passes when  $\theta = \delta/2$  and instability both of the light and the photometer make it desirable to use a bracketing method for determining the minimum light position for the analyser, when using the Tardy method. The mean of two analyser readings above and below the null position and corresponding with equal galvanometer deflexions gives the true null reading. For the authors' equipment the best values of the bracket limits to minimize the effect of instability on the analyser readings have been calculated to be changes of analyser reading of  $\pm 5^\circ$ .

When the behaviour of the photometer is satisfactory, attention is turned to the polariscope. Examination over different parts of the field showed that no deviation in the plane of polarization could be detected. The quarter-wave plates are open to more suspicion as, on the authors' bench, a departure from quadrature of about  $5^\circ$  in the same sense was found in each plate. It can be shown that, for the model retardation to be measured by the Tardy method to within  $1/500$  fringe, the errors in the quarter-wave plates should not be more than  $7^\circ$ . If the actual retardations of the quarter-wave plates were known, experimental results could be corrected by using a calibration curve.

#### ACKNOWLEDGMENT

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## APPENDIX

Mention has been made of the high degree of stability needed in the light source in order to be able to realize the high precision inherent in the photometer. Battery operation has proved successful but is cumbersome as four separate supplies are needed. The Electronics

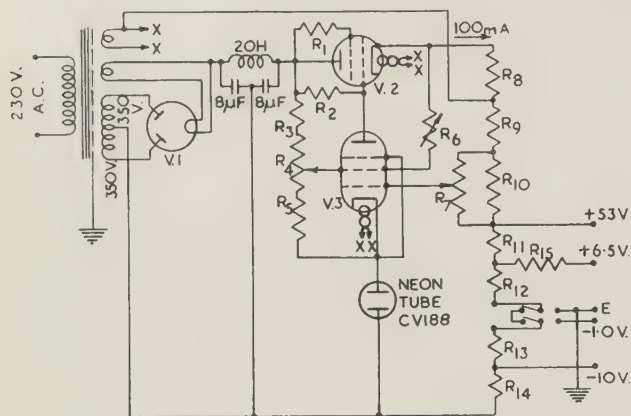


Fig. 11. Circuit diagram of stabilized power supply

$R_1$ , 100  $\Omega$  0.25 W;  $R_2$ , 1 M $\Omega$  0.5 W;  $R_3$ , 53.3 k $\Omega$  0.25 W;  $R_4$ , 5 k $\Omega$  potentiometer;  $R_5$ , 22 k $\Omega$  0.25 W;  $R_6$ , 100 k $\Omega$  variable;  $R_7$ , 2 k $\Omega$  potentiometer;  $R_8$ ,  $R_{10}$ ,  $R_{14}$ , 200  $\Omega$ ;  $R_9$ , 300  $\Omega$ ;  $R_{11}$ , 500  $\Omega$ ;  $R_{12}$ ,  $R_{13}$ , 100  $\Omega$ ;  $R_{15}$ , 1 k $\Omega$  0.5 W ( $R_8$ – $R_{14}$  are made up from 100  $\Omega$  2 W units);  $V_1$ , Osram U50;  $V_2$ , Osram KT66;  $V_3$ , Mullard EF36.

Section of the Division was consulted on the possibility of a mains-operated supply and, after some development work, a satisfactory solution was found.

Fig. 11 gives the circuit used and includes the values of the various components. Control is exercised by

varying the d.c. resistance of the valve  $V_2$  (KT66) so that the voltage across the cathode load ( $R_8 - R_{14}$ ), and hence the current through it, is maintained constant irrespective of mains voltage variations. The control signal which is applied to the grid of  $V_2$  is obtained from the valve  $V_3$  (EF36) and is initiated by the fluctuations of the potential difference between the screen grid and the cathode of this valve as the input voltage varies. The mean value of the current which is to flow through  $V_2$  is determined by the grid-cathode potential of  $V_3$  and is adjusted by means of  $R_7$ . The resistances  $R_3$ ,  $R_4$ ,  $R_5$  and  $R_6$  form a potential divider by means of which a proportion of the input voltage fluctuations can be applied to the screen grid of  $V_3$ . This proportion, which is determined by the setting of  $R_6$ , is adjusted so that there is minimum disturbance of the output voltage by the input voltage fluctuations.

The initial balancing of the system is carried out as follows.  $R_6$  is disconnected and  $R_7$  is adjusted for correct current flow through the electrometer triode filament (for this purpose, a switch is provided to replace the filament by a dummy resistance during balancing).  $R_4$  is then adjusted until there is zero potential difference between the cathode of  $V_2$  and the screen grid of  $V_3$ .  $R_6$  is re-connected and adjusted for minimum ripple in the output voltage (for this purpose, an artificial ripple can be injected into the input voltage by means of a small heater transformer in series, the output from the circuit being observed with an oscilloscope).

The degree of stabilization was measured by changing the mains voltage by  $\pm 15$  in 230 V. The resulting change in output voltage was less than 0.001 V in 200 V, a ratio of 1 : 25 000, and this performance was quite satisfactory for the operation of the photometer.

## The Determination of the Friction Couple in Balanced Rotating Mechanisms

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[Paper received 3 August, 1949]

A stroboscopic method is described which uses a lamp flashing at a fixed frequency, for the determination of the deceleration of balanced rotating mechanisms. From a knowledge of the moment of inertia of the mechanism, the friction couple, as a function of the speed, can be determined. The application of the method to the determination of the friction couple in a simple ballrace mechanism is described and some further applications are suggested. The method requires only the addition of a light stroboscopic disk to the mechanism.

A stroboscopic method is described for obtaining the angular-velocity-time curve for a freely rotating mechanism subjected to the retarding couple of its bearings. From such a curve the angular deceleration-angular velocity curve may be derived and, if the moment of inertia of the rotating system is calculable, a graph of the friction couple as a function of the angular velocity may be obtained. An application of the method is in the measurement of the friction couple in a ball-race

system for two loads over a range of speeds from 6000 to zero r.p.m. The stroboscopic lamp flashes at a constant frequency and illuminates the stroboscopic disk, Fig. 2, which is attached to the rotating system. As the latter decelerates, a succession of stationary patterns are observed and their occurrence recorded on a chronograph. The merit of the method is that it requires only the addition of a light stroboscopic disk to the rotating system.



## DETAILS OF THE MECHANISM TESTED

A section of the mechanism used as a test of the method is shown in Fig. 1. The brass disk *A*, approximately 4 in. diameter and  $\frac{1}{4}$  in. thick, is attached to the shaft *B* which, in turn, is supported by the ball-races *C* and *D*. These are standard seven-ball races by S.K.F. Co.,  $\frac{7}{8}$  in. external diameter and  $\frac{3}{8}$  in. internal diameter. No special precautions were taken in selecting the races, nor was the investigation extended to include the effect of lubrication on the friction couple. The lubricant used was thin machine oil. The load was varied by adding an additional disk on the shaft. Since

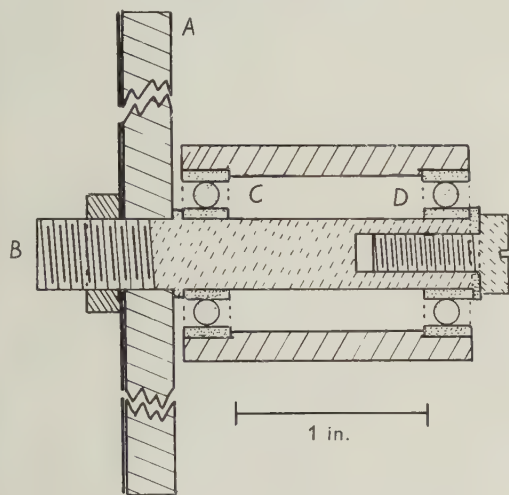


Fig. 1. Section of the mechanism

this varies the position of the centre of gravity of the moving system, the ratio of the loads on each ball-race is not constant under different total loads. The effect of this is, however, small except when the friction couple varies considerably with the load.

## THE STROBOSCOPIC APPARATUS AND METHOD

The disk is shown in Fig. 2. Patterns of 1, 6, 7, 8, 9, 10 and 11 spots were used, the even numbered spots being joined to give a 3, 4, and 5-spot pattern. Such an arrangement gives a large number of stationary patterns between zero speed and that at which the 1-spot pattern is stationary. The sequence of stationary patterns may, in the earlier stages of a run, occur too close together, but can be recorded when the mechanism has slowed down. In addition, stationary patterns can be recorded for the doubling of the number of spots as a momentarily stationary pattern when the mechanism is approaching its rest position.

For speeds of 1000 r.p.m. and less, a "beehive" neon lamp energized by the 50 c/s mains was found suitable. This source gives 6000 flashes per min. but the ratio of the duration of the flash to the duration of extinction is too large for speeds of the order of 1000 r.p.m., i.e. with the 6-spot pattern stationary. In investigating speeds up to 6000 r.p.m. it was found convenient to use a hot-

cathode mercury discharge lamp, the flash being produced by discharging a  $30 \mu\text{F}$  condenser, charged to 24 V., through the lamp. The charge and discharge of the condenser was effected by a contact attached to one of the prongs of an electrically maintained tuning fork of frequency 100 c/s. Even so, the duration of the flash, although much better than the flash with the neon lamp, made the stationary patterns difficult to observe at high speeds and a stroboson lamp, controlled by the a.c. mains or by a tuning fork, would have been better. Improvements in the shortness of the flash add to the ease of observation rather than to the accuracy of the experiment.

The interval between successive stationary patterns can be measured with a stop-watch, but this necessitates a

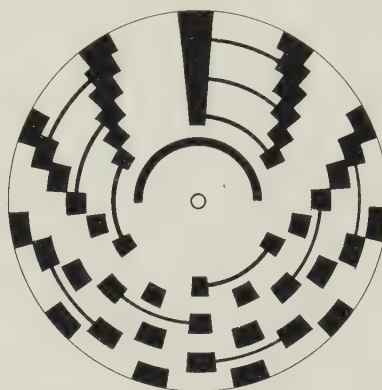


Fig. 2. Stroboscopic disk

large number of runs where a large number of stationary patterns is involved. In the investigation described, the time intervals were recorded on a hand-driven, smoked-drum chronograph by a style attached to the moving element of a Post Office relay. This relay was energized every 0.3 sec. by a commutator attached to an electric clock motor, the commutator discharging a  $4 \mu\text{F}$  condenser connected by a suitable resistance to a 60 V. h.t. battery. The value of the resistance was chosen so that the relay was not energized by the current passing through the resistance. Recording of successive stationary patterns was done by a tapping key which discharged a  $4 \mu\text{F}$  condenser through a second relay fitted with a style to mark the drum. Care was taken to adjust the positions of the two styles so that the synchronization mark on the drum was close to the corresponding time-mark.

The disk was set rotating at a speed slightly greater than 6000 r.p.m. by a hand-held electric motor fitted with a Bakelite fabric disk. This disk was pressed against the brass disk of the mechanism and moved radially towards the centre until the desired speed was attained. The assistant operating the chronograph then started turning and the observer operated the tapping key whenever a stationary pattern was observed. As there are about 30 stationary patterns between 6000 and 1000 r.p.m. some may be missed, so the services of a second assistant

are desirable to take down the sequence of stationary patterns called out by the observer as he presses the tapping key. This is of great assistance in the interpretation of the chronograph record at the end of a run. The sequence varied slightly in successive runs but was generally as follows: 1, 10, 5, 8, etc., corresponding to fractions of  $1, \frac{1}{10}, \frac{4}{5}, \frac{3}{4}$ , etc., of the flashing frequency of 6 000 per min.

#### INTERPRETATION OF THE CHRONOGRAPH RECORDS

From the chronograph record, a table of fraction of flashing frequency and time (from the 1-spot pattern stationary) was drawn up, the time being estimated to 0.06 sec. By taking successive differences, a table giving the values "differences of speed divided by the corresponding differences of time" (i.e. the average deceleration) and the average speed over the interval was obtained. Calculation of the moment of inertia of the rotating disk yields the appropriate numerical factor for converting the deceleration into the frictional couple. It will be noted that it is the average deceleration which was calculated. A refinement would be to make a smoothed (velocity-time) graph and find the slope at various points along it. It is unlikely, however, that the magnitude of the friction couple is required to a degree of accuracy to warrant this more laborious method of computation.

#### RESULTS OF THE INVESTIGATION

Fig. 3 shows the couple-speed graphs for total loads of 414 gm. and 841 gm. It will be seen that there is a small but measurable increase in the friction couple with load. In addition, points corresponding to different runs with the same load have been distinguished on the graphs showing that the performance of the mechanism was consistent and the curves reproducible.

The purpose of the investigation described above was

not specifically to determine the friction in ball-races but to investigate the applicability of the stroboscopic method for determining generally the friction couple in balanced rotating mechanisms. It could be applied, for example, to journal bearings under different loads and with different lubricants, to gear trains or to gyroscope

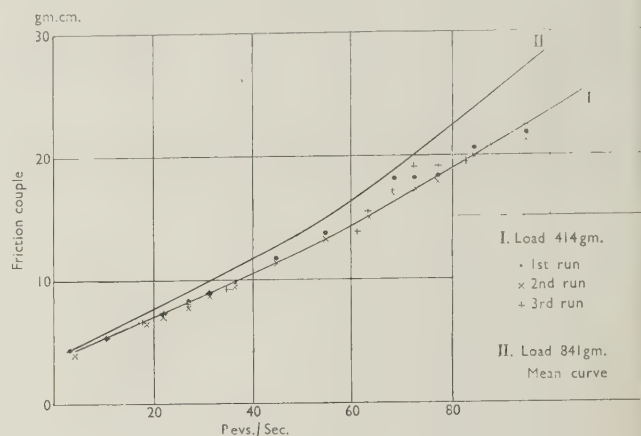


Fig. 3. Couple-speed graphs

and electric motor rotors. In addition, it could be used in calculating the power required to drive the mechanism at a given speed. Since the method requires only the addition of a light stroboscopic disk to the mechanism, the method of measurement will have a negligible effect on the performance of the mechanism. This is not the case if some forms of tachometer are used. If the angular deceleration is large, the time intervals between successive stationary patterns are small and the personal error of the observer in pressing the tapping key becomes an increasingly large fraction of the time interval, with consequent errors in determining the deceleration. This sets a limit on the applicability of the method.

## Gloss Measurement of Papers: Application of the Barkas Analysis

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[Paper first received 23 June, 1949 and in final form 18 July, 1949]

The Barkas analysis is applied to a series of nine papers of known visual gloss. With some reservations, the analysis is found to hold approximately and the distribution of mirror facets so derived is found to accord well with the surface structure of the papers as found by microscopic examination. It is concluded that, whilst the analysis is of negligible value for practical gloss determinations, it appears to present a useful method of investigating the surface structure and "finish" of paper and similar delicate materials. The gloss of the papers examined appears to be assessed solely by the intensity of the light reflected at the specular angle and not by the contrast noted by examination at different angles nor by the ratio of specularly-reflected to diffusely-reflected light. Some suggestions for future research are given.

Gloss measurement of papers is a problem of some importance in the printing and papermaking trades, since the appearance of the finished print depends upon, amongst other factors, the gloss and smoothness of the

paper supporting the ink film. The several different methods so far suggested for measuring gloss not only give widely different numerical results, but even grade papers in different orders. Precise information on the



correlation existing between visual assessments of gloss and instrumental gradings is meagre, and the present paper is the third in a series attempting to provide such information. In the summer of 1946 a questionnaire was circulated to printers, papermakers and some others known to be interested in gloss measurement. Each of those approached was asked to grade by eye a set of papers in order of gloss. The results of this assessment have now been compared with gradings of the same papers according to the Ingersoll glarimeter (1), the Bausch and Lomb and Sheen glossmeters, the Patra-Hilger goniophotometer and the Jones, Askania, Goerz, Ostwald and Klughardt conventions (2). It has been shown that of all the methods so far examined, the best results are given by direct measurement of the intensity of the light "specularly reflected" at an angle of  $45^\circ$  to the surface—though this need not necessarily be true of papers lying outside the comparatively narrow range of gloss tested.

Many workers in the past have realized that the only complete physical specification of the light-reflecting properties of a surface under set conditions of illumination is afforded by goniophotometric measurements, usually expressed in the form of polar curves showing the intensity of the light scattered at different angles to the test surface. There is no difficulty about determining such curves to any reasonable degree of accuracy: the real difficulty is to know what to do with them when obtained. Unless curves can be specified in terms of one or two simple parameters, which in this case is not usually so, they become awkward to handle; neither is it always easy to tell by inspection of polar curves which of the surfaces to which they correspond will appear the most glossy to a normal observer. In the great majority of cases it is quite impossible by any simple means to separate specular and diffuse components in the reflected light, if indeed they exist. As far as I have been able to discover, the only attempt at a scientific analysis of such curves is that of Barkas (3), who has also been the first to show clearly that the specularly-reflecting elements in the surface are not necessarily confined to the mean plane of the paper but may be oriented over a range of angles. Barkas proceeds on the assumption that the surface examined can be replaced by an optically-equivalent surface made up of specularly-reflecting and diffusely-reflecting facets. Both kinds of facet may be oriented at any angle to the mean surface; the specularly-reflecting ("mirror") facets are assumed to obey Fresnel's law, the diffusely-reflecting ("rough") to obey Lambert's law. The optical properties of the surface are expressed in terms of the relative areas and orientations of the two kinds of facet. It is not suggested that the facets necessarily have a physical existence within the surface, but if an optically-equivalent surface can be found consisting of such facets, then their areas and distributions will provide parameters defining the optical behaviour of the real surface under examination. In the case of paper facets approximating to the specularly-reflecting

facets of the analysis certainly exist, as will be shown later: there is, strictly, no such thing as a diffusely-reflecting facet, but the concept provides a useful way of summarizing the effects of repeated reflexions and refractions at the fibre-air interfaces within the sheet.

On this basis Barkas develops the fundamental equation

$$E_{\alpha\beta} \cos \alpha = \frac{1}{2}rA[\cos(\alpha - \beta) + \cos(\alpha + \beta)] + sB \cos \frac{1}{2}(\alpha - \beta)$$

where  $E_{\alpha\beta}$  is the total flux received from the surface illuminated at angle  $\alpha$  and viewed at angle  $\beta$ .  $A$  is the relative area of rough facets illuminated under these conditions and  $r$  the proportion of incident flux scattered normally to a rough facet: in practice  $rA$  is treated as a single factor defining the diffuse component of the scattered flux.  $B$  is the relative area of mirror facets at an angle  $\frac{1}{2}(\alpha + \beta)$  to the mean surface and  $s$  is the proportion of incident flux reflected at the specular angle from a mirror facet.  $E_{\alpha\beta}$  is measured experimentally and  $s$  is calculated from Fresnel's equation. From these data  $rA$  and  $B$  (both of which are functions of  $\alpha$  and  $\beta$ ) can be determined by a method outlined in Barkas's paper. It is evident that, if true, the Barkas equation provides a powerful tool for the investigation of semi-glossy surfaces. It relates the otherwise apparently unconnected polar curves for different angles of incidence, replaces arbitrary "gloss numbers" by factors having a definite relation to the physical properties of the reflecting surface and, thirdly, it should give information about the structure of paper without injury to its surface. For these reasons, the Barkas analysis has been applied to the already mentioned set of nine papers of known relative visual gloss with the object of ascertaining whether the analysis applies to these papers and, if so, whether the factors  $rA$  and  $B$  bear any relation to the visual gloss and surface structure of the papers.

#### THE PAPERS

The papers used in this investigation (numbered  $A_1$ – $A_9$ ) were war-grade printings of moderate reflexion factor and varying in gloss from almost matt ( $A_8$ ) to a "super-calendered" finish ( $A_5$ ). An analysis of the papers is given, since it will be necessary to refer to it later. The traces of rag, straw, etc., mentioned have undoubtedly been introduced through the use of waste and salvaged paper in addition to new pulp. This was unavoidable at the time these papers were manufactured. The ash in all cases consists mainly of mineral loading in the form of china clay.

Papers  $A_1$ ,  $A_2$ ,  $A_4$ ,  $A_5$ ,  $A_7$  and  $A_8$

Furnish: 70–80% bleached coniferous thin wood (probably bleached sulphite pulp), the remainder chiefly a grass or straw fibre, together with some deciduous wood and traces of mechanical wood, rag and unbleached sulphite, the whole very much cut up by beating.

Ash:	A <sub>1</sub>	A <sub>2</sub>	A <sub>4</sub>	A <sub>5</sub>	A <sub>7</sub>	A <sub>8</sub>
	11.6%	8.8%	1.6%	23.5%	15.0%	3.2%

Paper A<sub>3</sub>

Furnish: 65% coniferous chemical wood (probably sulphite and either lightly bleached or very well cooked); 35% mechanical wood; traces of straw and deciduous wood.

Ash: 13.9%.

Paper A<sub>6</sub>

Furnish: 65% mechanical wood; 20% unbleached sulphite pulp; 15% bleached chemical wood (probably sulphite); traces of straw and deciduous wood.

Ash: 15.0%.

Paper A<sub>9</sub>

Furnish: Bleached coniferous chemical wood (probably sulphite with a small amount of sulphate); trace of deciduous wood.

Ash: 16.0%.

The gloss of these papers was imparted by "calendering." Coming straight from the machine, paper has an almost matt surface. The gloss is produced by damping the paper and passing it through a stack of polished steel rollers under more or less pressure, according to the degree of gloss required. The calendering presses the fibres together, making the surface smoother, more shiny and compact, but it also reduces the reflexion factor and opacity. The gloss produced by calendering is never quite uniform, since slight variations in the thickness of the web are impossible to avoid and the thicker parts, receiving more pressure, are the more highly glazed. The visual gloss of the papers on an arbitrary psychological scale ranging from 0 to 10 is

A <sub>8</sub>	A <sub>4</sub>	A <sub>1</sub>	A <sub>3</sub>	A <sub>9</sub>	A <sub>6</sub>	A <sub>7</sub>	A <sub>2</sub>	A <sub>5</sub>
0.43	1.14	2.98	3.70	5.72	7.75	8.22	8.87	9.34

## EXPERIMENTAL RESULTS

The goniophotometric measurements on which this paper is based were made with the Patra-Hilger goniophotometer (4). Light from a 6 V. 18 A. tungsten ribbon-filament projector lamp run at its rated voltage was filtered through "Calorex" Chance glass to reduce infra-red radiation. The photocell employed had maximum sensitivity at 550  $\mu\mu$  falling off to zero at 400  $\mu\mu$  and 760  $\mu\mu$ . The incident and reflected beams were in the form of thin wedges of total angular aperture 2° 12' and the part of specimen illuminated was approximately 10 mm. long and 2 mm. wide. Specimens were mounted with "top sides" facing the light and "cross directions" parallel to the long side of the illuminated patch. These measurements are available to anyone interested and arrangements are being made to have them reproduced in full elsewhere. Shortage of space forbids their in-

clusion here, but Figs. 1 and 2 are the polar curves for the extreme papers of the series, A<sub>8</sub> and A<sub>5</sub>, taken at 15° intervals and illustrating the types of curve obtained. The intensities of light received by the photocell are in all cases expressed as a percentage of the intensity received by the cell when placed normally to a surface of freshly prepared magnesium oxide illuminated at 45° by the same incident beam; they are thus directly comparable. The accuracy of measurement is such that the

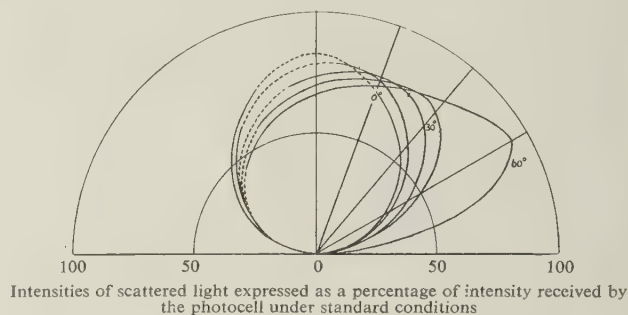


Fig. 1. Polar curves for paper A<sub>8</sub> (machine-finish)

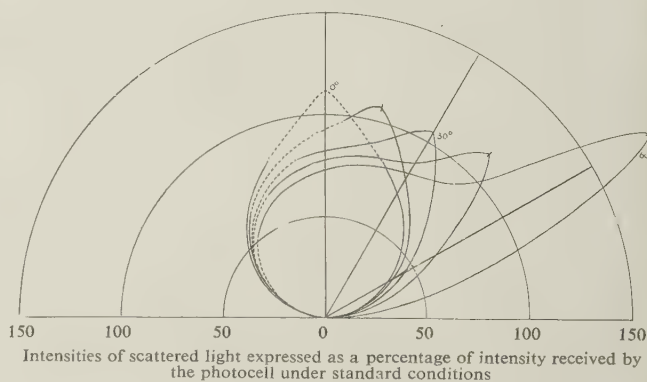


Fig. 2. Polar curves for paper A<sub>5</sub> (super-calendered)

recorded intensities will be within 1% (or  $\frac{1}{2}$  unit—which ever is the greater) of the true intensities in 19 cases out of 20. Measurements have been made over as wide a range of angles of incidence and viewing as possible, but the design of the goniophotometer does not at present permit of observations with  $\alpha$  or  $\beta$  greater than 75°, nor with  $(\alpha - \beta)$  less than 20°. The same part of the specimen was illuminated throughout the entire series of observations. The effect of sampling variations will be discussed in the next section.

## EFFECT OF SAMPLING VARIATIONS

Sampling variations are the bugbear of all physical testing of paper. It is impossible to make a perfectly uniform ream of paper and hence one is usually obliged to make measurements on a number of samples taken from the ream or roll and to treat the results statistically. The measurements to which reference is made are valid in each case for a strip of paper 10 mm. long and 2 mm.



wide. Since the same small strip has been illuminated at all the recorded angles of incidence and viewing, the results of the analysis are applicable without reservation to this strip. It is, however, a great assumption to take this as representative of the paper as a whole: indeed, if the small strip in question contained a large shive (piece of un-disintegrated wood) or flaw, the results could be altogether misleading.

Ideally, the data should be repeated on at least 19 more samples of each paper and the analysis carried out independently on each sample, so that the effects of sampling variations on the values of  $rA$  and  $B$  could be studied. Unfortunately the labour required to make even the present limited sets of measurements and the computations from them has been considerable and the work required to complete the ideal programme could not be done within any reasonable length of time without automatic methods of recording and computation. It has been possible here only to ensure that the

forecast, but in general an increase near the specular angle will increase  $B$  by a roughly proportionate amount and will cause a decrease in  $rA$ .

#### ANALYSIS

The measurements plotted in the appropriate manner [ $E_{\alpha\beta} \cos \alpha$  against  $\cos(\alpha - \beta)$  for  $(\alpha + \beta)$  constant] give families of curves closely similar, though differing in some details, to those published by Barkas and two points merit discussion. Firstly, the Barkas equation is symmetrical with respect to  $\alpha$  and  $\beta$  and one will therefore expect to find  $E_{\alpha\beta} \cos \alpha = E_{\beta\alpha} \cos \beta$  for a given numerical value of  $(\alpha + \beta)$ . Within the limits of experimental error, this is so in almost all cases for  $(\alpha + \beta) > 30^\circ$ . It is also true for  $(\alpha + \beta) \geq 30^\circ$  for papers  $A_1$ ,  $A_4$ ,  $A_7$  and  $A_8$ , but it is not true under these conditions for  $A_5$  and  $A_6$  and there are indications that it is not true for  $A_2$ ,  $A_3$  and  $A_9$ . As an illustration figures are given for  $A_6$  for  $(\alpha + \beta) = \pm 10^\circ$ .

Table 1. Values of  $E_{\alpha\beta} \cos \alpha$  for paper  $A_6$ ,  $(\alpha + \beta) = \pm 10^\circ$

$\alpha - \beta$	20	30	40	50	60	70	80	90	100	110	120	130	140
$E_{\alpha\beta} \cos \alpha$ {	79	75½	73½	69	66	64	60½	57	54½	54	53	55	65
	77	74	70½	68½	64	61½	57	53½	51½	51	51	50½	56

recorded data are neither freak results nor seriously uncharacteristic of the reams to which they are attributed, but owing to lack of proper statistical data, it is impossible to judge conclusively how far small differences in the derived values of  $rA$  and  $B$  are significant or not and only broad conclusions can be drawn. This is less serious than might at first sight be expected, for there are at present many uncertainties in the Barkas analysis itself and much research will have to be completed before it is possible to deal with nice differences in the values of the factors.

As a check on the values complete polar curves at  $45^\circ$  and  $60^\circ$  were determined for an additional four samples of each paper. They will be made available elsewhere with the other data. Inspection of these curves showed that the intensities already measured were almost certainly not freak values, though some sampling variations were clearly revealed. Most of the variations occurred near the angle of specular reflexion, though there were also certain small variations in the shapes of the curves. It was noticed further that the papers of lowest gloss ( $A_4$ ,  $A_8$ ,  $A_1$ ,  $A_3$ ) showed comparatively small variations, whereas the calendered papers ( $A_9$ ,  $A_6$ ,  $A_7$ ,  $A_2$ ,  $A_5$ ) showed greater variations as the gloss increased: this accords well with the tendencies noticed in previous work (see Table 2, ref. 2). As stated, these data were inadequate for statistical treatment but it seemed not unreasonable to assume from them that the main measurements were not freak results due to the illumination of shives or flaws and were within 5% of the true means in most cases (less with the machine-finished papers). The effect of sampling variations on the values of  $rA$  and  $B$  in the analysis cannot easily be

The upper line for  $E_{\alpha\beta} \cos \alpha$  is for angle of incidence numerically greater than angle of viewing; the lower line for angle of incidence numerically less than angle of viewing. There is a small, but systematic, difference between the two sets which increases as  $(\alpha - \beta)$  increases. For the calculations of  $rA$  and  $B$  recorded later, this difference was ignored and the mean value taken, but it is evident that the curve really consists of two branches. There is apparently nothing abnormal in the structure of the paper for the polar curve for  $\alpha = 0$  is symmetrical with respect to  $\beta$ .

Barkas has drawn the author's attention to the fact that where a difference between the two sets of values does exist, the greater value in each pair is almost invariably for an angle of incidence numerically greater than the angle of viewing. This seems to be quite general and it does not appear to be due to instrumental errors since exactly the same tendency can be discovered in the original data on which his 1939 paper was founded, such data having been obtained with a goniophotometer of entirely different construction. Careful examination of the data has shown that, in some cases, the Helmholtz reciprocity law is not strictly valid. It holds within the accuracy of measurement for most of the papers but there are nevertheless some significant discrepancies. Thus for paper  $A_6$  the luminance (intensity/cosine of angle of viewing) for  $45^\circ$  incidence and  $0^\circ$  viewing is  $62\frac{1}{2}$ , whereas for  $0^\circ$  incidence and  $45^\circ$  viewing it is only 60; for  $20^\circ$  incidence and  $0^\circ$  viewing it is 72, for  $0^\circ$  incidence and  $20^\circ$  viewing only 70. The differences are quite systematic. Similar, though smaller, differences are shown by  $A_9$ . On the other hand, for paper  $A_3$ , the reciprocity law holds well within the accuracy of measurement.

This point is of some importance in view of a current tendency to change the standard C.I.E. conditions of illuminating and viewing specimens from 45° incidence and normal viewing to normal incidence and 45° viewing: these conditions are not necessarily precisely equivalent.

Secondly, the exact course of the curves for  $(\alpha + \beta) > 60^\circ$  has been found difficult to determine owing to lack of data when  $\alpha > 75^\circ$ . According to the Barkas equation, the curves degenerate into straight lines as soon as mirror components become negligible [usually at  $(\alpha + \beta) = 60^\circ$  or  $70^\circ$ ] and  $E_{\alpha\beta} \cos \alpha$  becomes zero as  $\alpha \rightarrow 90^\circ$ . Consequently all curves past  $(\alpha + \beta) = 60^\circ$  should be straight lines cutting the  $\cos(\alpha - \beta)$  axis at the point corresponding to  $\alpha = 90^\circ$ . Fig. 3 shows some

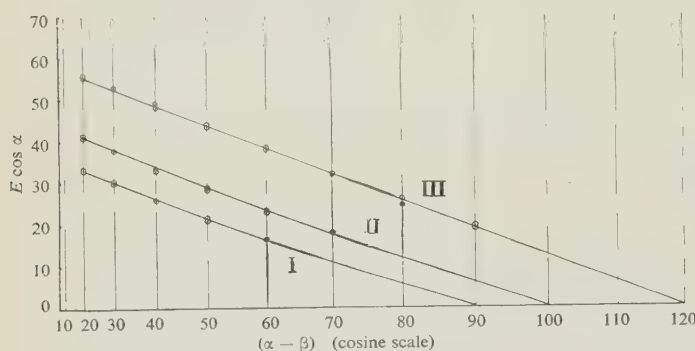


Fig. 3. Some experimental points obtained on paper A<sub>1</sub>  
Values of  $(\alpha + \beta)$ : Curve I,  $90^\circ$ ; II,  $80^\circ$ ; III,  $60^\circ$

of the experimental points obtained with paper A<sub>1</sub>: the results are typical and are shown broadly by all the other papers, though numerical values are different. In examining these, it should be borne in mind that the actual values of  $E_{\alpha\beta}$  corresponding to the pairs of points are in many cases widely different, though values of  $E_{\alpha\beta} \cos \alpha$  work out closely the same. With  $(\alpha + \beta) = 60^\circ$  the experimental points are best represented by a straight line cutting the axis at  $\cos(\alpha - \beta) = -0.5$ , exactly as required by the Barkas equation when  $B = 0$ . For  $(\alpha + \beta) = 80^\circ$  and  $(\alpha + \beta) = 90^\circ$ , however, the points are best represented as shown, by curves cutting the

axis at the appropriate points. The Barkas equation does not allow for the development of curves once  $B$  reaches zero (unless by making the unpalatable assumption that  $B$ , having decreased steadily to zero as  $(\alpha + \beta)$  reaches  $60^\circ$ , becomes positive again for larger values of  $(\alpha + \beta)$ ; nor is it easy to see the physical significance of the curvature, which, if genuine, makes the estimation of  $rA$  impossible for large values of  $(\alpha + \beta)$ . Alternatively, it is possible to draw straight lines through the experimental points for  $(\alpha + \beta) = 80^\circ$  and  $90^\circ$  without doing too much violence to them; but the straight lines cut the axis at the wrong points. For example, a straight line drawn through the  $(\alpha + \beta) = 80^\circ$  set of points cuts the axis at approximately  $\cos(\alpha - \beta) = 0.09$ , which means that the scattered flux is zero when  $\alpha$  reaches about  $87\frac{1}{2}^\circ$  and  $\beta = 7\frac{1}{2}^\circ$ . This result is not acceptable although the flux is certainly small under such conditions.

In order to estimate values of  $\frac{1}{2}rA$  for large values of  $(\alpha + \beta)$ , it was decided to draw straight lines through the experimental points and calculate  $\frac{1}{2}rA$  from their slopes: the values of  $E_{\alpha\beta} \cos \alpha$  are then represented by an equation of the type:

$$(E_{\alpha\beta} \cos \alpha - e) = \frac{1}{2}(rA)[\cos(\alpha - \beta) + \cos(\alpha + \beta)]$$

where  $e$  is a small parameter dependent on  $(\alpha + \beta)$  and constant when  $(\alpha + \beta)$  is constant, which I have not been able to justify theoretically. Table 2 gives the values for  $\frac{1}{2}rA$  and  $e$  for various values of  $(\alpha + \beta)$ . If, as seems likely, the lines relating  $E_{\alpha\beta} \cos \alpha$  and  $\cos(\alpha - \beta)$  are curvilinear, the tabulated values of  $\frac{1}{2}rA$  will be too high: in any case, their values for  $(\alpha + \beta) > 60^\circ$  are very uncertain, for reasons discussed. Determination of the values of  $B$  involves the refractive index of the material composing the facets. Cellulose is doubly refracting and the refractive indices depend to some extent on the plant from which the fibres have been extracted. 1.56 is, however, a good mean value which happens to be that also of the china clay used as a filler in these papers. A mean refractive index of 1.56 has accordingly been adopted.

Table 3 gives typical values of  $B$  so determined. In

Table 2. Values of  $\frac{1}{2}rA$  and  $e$  at  $10^\circ$  intervals

$\alpha + \beta$	A <sub>1</sub>		A <sub>2</sub>		A <sub>3</sub>		A <sub>4</sub>		A <sub>5</sub>		A <sub>6</sub>		A <sub>7</sub>		A <sub>8</sub>		A <sub>9</sub>	
	$\frac{1}{2}rA$	$e$	$\frac{1}{2}rA$	$e$	$\frac{1}{2}rA$	$e$	$\frac{1}{2}rA$	$e$	$\frac{1}{2}rA$	$e$	$\frac{1}{2}rA$	$e$	$\frac{1}{2}rA$	$e$	$\frac{1}{2}rA$	$e$	$\frac{1}{2}rA$	$e$
0	27.9	—	18.3	—	21.4	—	27.9	—	17.6	—	0.8	—	18.9	—	22.0	—	24.3	—
10	30.7	—	28.8	—	23.3	—	29.7	—	26.0	—	19.3	—	25.6	—	22.8	—	30.1	—
20	33.6	—	32.5	—	26.2	—	32.2	—	33.7	—	23.3	—	32.4	—	24.3	—	34.7	—
30	35.7	—	35.2	—	30.8	—	35.0	—	36.2	—	26.0	—	34.9	—	27.1	—	36.6	—
40	38.8	—	36.6	—	31.5	—	36.8	—	37.7	—	27.9	—	36.4	—	29.9	—	38.1	—
50	39.4	—	36.6	—	32.0	—	37.3	—	38.3	0.0	28.6	—	37.2	0.0	31.3	—	39.1	—
60	38.9	0.4	37.0	0.6	32.7	0.0	37.9	0.0	38.4	2.3	29.2	0.9	36.9	0.5	31.8	—	39.3	0.4
70	38.7	1.8	37.6	3.0	33.4	2.7	38.4	1.6	38.8	4.0	30.3	3.0	37.7	2.4	31.4	0.0	39.0	2.3
80	39.0	2.5	38.1	4.0	34.3	3.5	37.1	1.8	39.3	5.2	30.2	3.6	37.2	2.8	32.3	2.0	38.4	3.2
90	38.5	3.2	37.6	3.7	33.6	3.3	37.0	2.5	38.5	5.0	30.0	4.0	37.6	4.0	32.0	2.3	39.2	4.0
100	37.5	2.2	38.0	4.2	33.8	4.0	36.5	2.7	39.2	4.9	30.2	4.0	39.7	5.3	32.2	2.6	38.5	4.0
110	36.5	2.4	37.6	4.8	34.5	3.8	36.0	2.0	39.5	5.0	30.8	3.8	38.0	4.7	31.5	2.7	39.0	3.4
120	37.0	2.0	41.7	4.8	36.3	4.5	36.3	1.9	40.5	4.8	30.8	2.8	40.0	4.4	32.5	2.6	39.0	3.3
130	37.0	1.6	42.3	4.5	38.1	4.7	36.5	1.9	41.5	3.9	31.0	2.6	42.0	3.6	31.5	2.3	39.0	3.2



Table 3. Typical values of  $B$  (the relative area of mirror facets) taking a mean refractive index of 1.56

$(\alpha - \beta)$	$(\alpha + \beta)$						
	0	10	20	30	40	50	60
0	14.05	6.95	3.9	1.85	0.9	0.4	0.0
10	14.1	6.95	3.9	1.85	0.85	0.4	0.0
20	14.0	6.95	3.9	1.9	0.85	0.4	0.0
30	14.1	7.0	3.9	1.9	0.85	0.4	0.0
40	14.05	7.05	3.9	1.95	0.85	0.45	0.0
50	14.0	7.1	3.9	2.0	0.95	0.35	0.0
60	14.0	7.15	3.9	2.05	0.95	0.35	0.0
70	14.0	7.15	3.9	2.1	1.0	0.35	0.0
80	14.0	7.2	3.9	2.15	1.0	0.4	0.0
90	13.9	7.2	3.9	2.15	1.0	0.4	0.0
100	13.85	7.2	3.8	2.1	1.0	0.4	
110	14.1	7.1	3.65	2.0	1.0		
120	14.5	7.1	3.4	1.8			
130	14.45	7.3	3.15				
140	18.7	8.1					
150	22.8						

studying the Table it will be found helpful to remember that  $\frac{1}{2}(\alpha + \beta)$  is the inclination of a facet to the mean plane of the paper and  $\frac{1}{2}(\alpha - \beta)$  is the angle of incidence on that facet. For a given value of  $(\alpha + \beta)$ , the values

Table 4. Mean values adopted for  $B$  (the relative area of mirror facets)

Paper	Visual gloss	$(\alpha + \beta)$							
		0	10	20	30	40	50	60	70
A <sub>5</sub>	9·34	15·16	8·78	3·86	1·96	0·75	0·00		
A <sub>2</sub>	8·87	14·02	7·07	3·90	1·99	0·92	0·39	0·00	
A <sub>7</sub>	8·22	14·05	8·57	4·12	2·18	1·00	0·24	0·00	
A <sub>6</sub>	7·75	19·12	8·60	4·86	2·71	1·30	0·56	0·00	
A <sub>9</sub>	5·72	11·84	7·66	4·41	2·64	1·38	0·42	0·00	
A <sub>3</sub>	3·70	10·32	8·03	5·36	2·66	1·53	0·73	0·00	
A <sub>1</sub>	2·98	8·08	6·24	4·25	2·59	0·96	0·18	0·00	
A <sub>4</sub>	1·14	6·93	5·94	4·50	2·70	1·34	0·68	0·00	
A <sub>8</sub>	0·43	7·56	6·71	5·37	3·52	1·86	0·85	0·20	0·00

of  $B$  are roughly independent of  $(\alpha - \beta)$  until  $(\alpha - \beta) = 120^\circ$ ; thereafter there is a rapid increase in the apparent values of  $B$  particularly with the glossier papers. The reason for this increase is not entirely clear, but may be due to the fact that at very oblique angles of incidence, corresponding to large values of  $(\alpha - \beta)$ , rough facets, like distempers for example, can become regularly reflecting. For example magnesium oxide on silver, which is as good a diffuser as any known reflecting surface, shows a distinct reddish specular component of intensity greater than would be predicted, from either Fresnel's laws or the Barkas analysis, when illuminated at  $75^\circ$  and over. An apparent increase in the areas of regularly reflecting facets at oblique angles of illumination thus seems quite plausible.

Barkas reports a progressive decrease in  $B$  with increasing values of  $(\alpha - \beta)$  and attributes this to gradual eclipsing of the mirror facets as the angle of incidence increases. The present figures show, however, that  $B$  is just as likely to increase as decrease over the range  $(\alpha - \beta) = 0$  to  $120^\circ$  and the most probable interpretation of the small fluctuations observed within this range is that they are due to errors in the determination of

$\frac{1}{2}rA$  or in choice of refractive index. In any case, with the papers employed, the incidence of masking can be observed through the microscope: it is quite inappreciable until  $\alpha$  exceeds  $50^\circ$ . For the purposes of this paper,  $B$  is taken as constant, for a given value of  $(\alpha + \beta)$ , and the "true" value is assumed to be the arithmetic mean of the recorded values over the range  $(\alpha - \beta) = 0$  to  $(\alpha - \beta) = 90^\circ$ . This procedure, though arbitrary, seems likely to give more representative values of  $B$  than those given for  $(\alpha - \beta) = 0$  alone. For  $(\alpha - \beta) > 120^\circ$  the Barkas equation breaks down.

## APPLICATION OF RESULTS

Table 4 gives the mean values of  $B$  which have been adopted, the papers being arranged in order of visual gloss with the most glossy at the top. Apart from A<sub>6</sub>, which is exceptional and will be discussed later, three salient features emerge. First,  $B$  [for  $(\alpha + \beta) = 0$ ] increases fairly steadily as the gloss increases. Second, as the gloss decreases so the relative area of facets inclined at the larger angles increases. Third, the distribution becomes more and more sharply "peaked" as the gloss increases. This is shown by Table 5 which

gives the standard deviation and kurtosis of each of the distributions given in Table 4.  $\sigma$  is the standard deviation calculated in the ordinary way and  $\gamma_2$  is defined as  $(\mu_4 - 3\mu_2^2)/\mu_2^2$  where  $\mu_2$  and  $\mu_4$  are the second and fourth moments of the distribution about the mean respectively.  $\gamma_2$  is zero for a normal distribution, negative for a platykurtic distribution and positive for a leptokurtic distribution.

Table 5. Standard deviation and kurtosis of each of the distributions given in Table 4

Paper	$\sigma$	$\gamma_2$
A <sub>5</sub>	1.53	+ 0.32
A <sub>2</sub>	1.75	+ 0.57
A <sub>7</sub>	1.70	+ 0.42
A <sub>6</sub>	1.78	+ 0.58
A <sub>9</sub>	1.91	+ 0.06
A <sub>3</sub>	2.02	- 0.06
A <sub>1</sub>	1.91	- 0.34
A <sub>4</sub>	2.16	- 0.36
A <sub>8</sub>	2.30	- 0.37

The general tendency of  $\sigma$  to decrease and the distribution to pass from platykurtic to leptokurtic with the increase of gloss is very clearly shown. In one

respect, the numerical values of  $\gamma_2$  are misleading. The fourth moment about the mean, on which  $\gamma_2$  depends, is very sensitive to values at large angles, even though small, and the omission of data for  $(\alpha + \beta) > 60^\circ$  or  $70^\circ$  in the tables tends to reduce the value of  $\gamma_2$ . In fact, the distribution of  $A_8$  is very nearly normal, as can be seen below, where the "calculated" figures apply to a normal distribution of the same standard deviation.

Table 6. Observed and calculated values of gloss

$\alpha + \beta$	0	10	20	30	40	50	60	70
Observed	7.56	6.71	5.37	3.52	1.86	0.85	0.20	0.00
Calculated	7.56	6.88	5.19	3.24	1.68	0.70	0.26	0.08

As the gloss increases, so does the tendency to assume the leptokurtic form.

All this is entirely consonant with what we know of the manufacture of the papers used in this investigation. The fibres of the machine-finish papers are as undisturbed as possible once they have left the Fourdrinier wire and the analysis gives us a plausible picture of a random distribution of reflecting surfaces tilted at all angles up to  $35^\circ$  to the mean surface. How reasonable this is is shown in Fig. 4 which is a stereophotomicro-

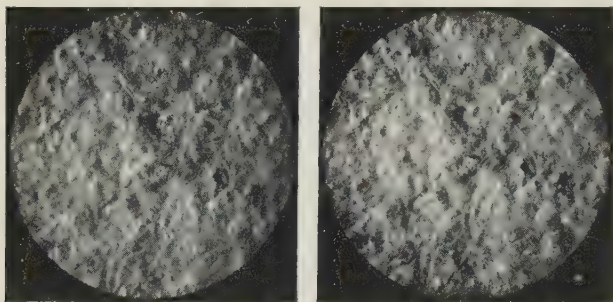


Fig. 4. Paper  $A_8$  illuminated at  $20^\circ$  to surface ( $\times 20$ )

graph of the surface of  $A_8$ , illuminated at  $70^\circ$  to the normal. The undulations of the surface and the numerous facets capable of reflecting light are clearly seen. As the paper is calendered, the fibres are forcibly persuaded to lie more and more in the mean plane of the paper and the area of reflecting facets in this plane is increased by burnishing. The result is shown in Fig. 5 showing paper  $A_5$  under the same conditions as

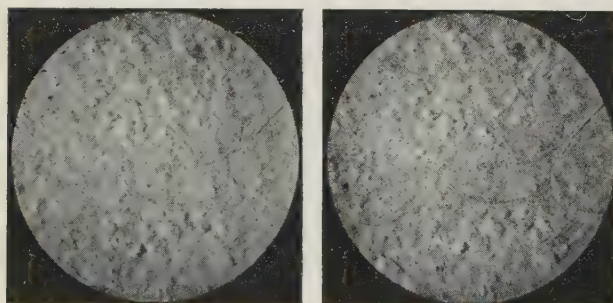


Fig. 5. Paper  $A_5$  illuminated at  $20^\circ$  to surface ( $\times 20$ )

Fig. 4. The surface is in striking contrast to that of the machine-finished paper. The reflecting surfaces are now mainly in the mean plane of the paper and those at other angles are confined to the small pits which have not yet been smoothed out by the calendering process. The extra mineral loading in  $A_5$  also helps to produce a smooth surface. The leptokurtic distribution of facets suggested by the analysis is therefore quite acceptable.

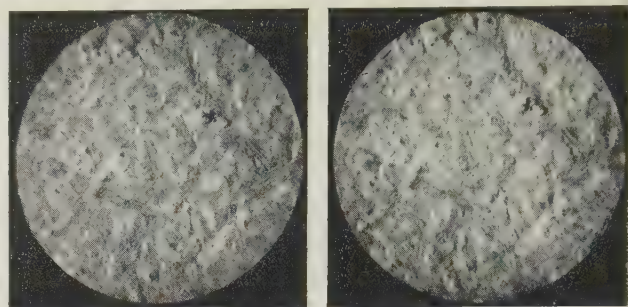


Fig. 6. Paper  $A_6$  illuminated at  $20^\circ$  to surface ( $\times 20$ )

It remains to discuss the exceptional paper  $A_6$ . For  $(\alpha + \beta) = 0$ , this has an unusually low value of  $rA$  and a high value of  $B$ , suggesting that most of the light received at the specular angle is, in fact, specularly reflected. Reference to the fibre furnish figures in this section "PAPERS" will show that paper  $A_6$  is exceptional in containing a large percentage of mechanical wood, which, as its name implies, is wood that has been mechanically disintegrated without any attempt to remove the lignin and tarry impurities which, in "chemical wood," are dissolved out by heating with sulphites, sulphides or other means. Chemical wood fibres are clear and fairly transparent. Mechanical wood fibres are always darker and more opaque, as is the paper made from them. Consequently  $A_6$  is of lower reflexion factor and greater opacity than any of the others. The microstructure is shown in Fig. 6, in which it is seen that the mechanical wood occurs in the form of comparatively large opaque plates which are more or less parallel to the plane of the mean surface. The exceptionally high value of  $B$  for  $(\alpha + \beta) = 0$  is thereby explained. Similarly, the low

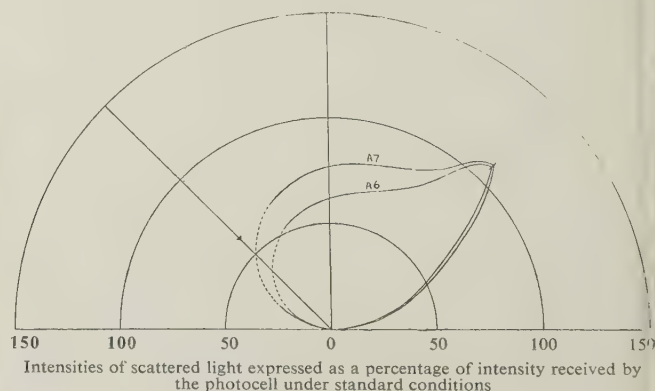


Fig. 7. Polar curves for papers  $A_6$  and  $A_7$



values of  $rA$  are due to the opacity of the mechanical wood, which reduces the number of possible reflexions and refractions at fibre-air interfaces. The Barkas analysis therefore gives a true description of the properties of the surface. It is, however, an interesting, and to the author surprising, fact that  $A_6$  does not appear the glossiest of the series. Fig. 7 shows the polar curves for papers  $A_6$  and  $A_7$  at an angle of incidence of  $45^\circ$ . The curves suggest, and the analysis confirms, that the ratio of specularly reflected to diffusely reflected light is greater with  $A_6$  than with  $A_7$ ; but  $A_7$  is ranked the higher by most observers (1). It appears that the gloss of this series of papers is judged mainly by the intensity of the light emitted at the specular angle, not by the contrast noted at various angles of viewing, nor by the ratio of specularly reflected to diffusely-reflected light.

### CONCLUSION

For practical gloss determinations, the Barkas analysis is of little value. Though the values of  $B$  are correlated with the visual gloss of the surface, particularly when papers of the same furnish are being compared, the labour needed to establish them is out of all proportion to their value. More reliable gloss specifications may be obtained in literally a thousandth of the time by measurement of the intensity of the light reflected at the specular angle when the angle of incidence is  $45^\circ$ . On the other hand the analysis does account qualitatively, and in many cases quantitatively, for the shape of the polar reflexion curves and relates them in a way otherwise impossible. More important, however, is the possible application of the method for exploring the surface structure of papers and other delicate materials. The surface contour of paper is of great importance in practical printing and any method of determining this contour without damaging the surface deserves close examination. Whilst the Barkas analysis alone cannot provide

a complete answer to this problem, it appears to offer information which may prove valuable. A few hints to possible future investigators in this field may not be out of place. The form of the curves for  $(\alpha + \beta) > 60^\circ$  should be established more precisely than has here been possible. This means the construction of a gonio-photometer capable of operating over a wider range of angles of incidence and viewing than the instrument at the disposal of the author. Particular attention should be given to angles of incidence greater than  $75^\circ$ . On the other hand, those only interested in the analysis as a possible means of investigating surface structure may confine their attention to the curves  $(\alpha + \beta) = 0^\circ$  to  $(\alpha + \beta) = 60^\circ$  over the range  $(\alpha - \beta) = 20^\circ$  to  $(\alpha - \beta) = 120^\circ$  which will reduce the number of observations to be made. For precise work, it will be essential to obtain data sufficient to permit of statistical treatment. If such data are required within any reasonable time, the use of the geared turntable described by Barkas (5) together with automatic means of rotating it and recording the appropriate intensities will be found unavoidable. Even so the amount of computation required will be considerable, though experience may show how simplifications may be made without loss of accuracy.

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## The Use of Cold Cathode Relay Valves with Grid-Cathode Circuits of High Resistance

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The grid current drawn by a cold-cathode relay valve near the critical grid potential normally precludes the use of a valve of this type where the grid circuit contains a high series resistance. By superimposing a train of voltage pulses on the applied grid potential this limitation may be removed. The instantaneous grid current may then be high whilst the mean value is much less. The limiting resistance usable in the grid circuit is thereby increased a hundredfold or more. In many cases the pulses may conveniently be derived from the rectifier ripple.

Several practical applications of the principle are described.

For many purposes the abrupt change of anode current with bias voltage of the gas-filled triode is preferable to the gradual change which is characteristic of the hard valve. The recently introduced cold-cathode gas-filled valves offer several advantages over the hot-cathode types. They trigger at a positive bias, the value

of which is for some types substantially independent of the anode voltage. Frequently the glow from the cathode may be used to indicate when the valve is conducting. Unfortunately, as the grid potential approaches the point at which the tube "fires," the grid current increases rapidly to comparatively large values and, in

certain circumstances, this effect seriously limits the maximum value of resistance which can be included in the grid circuit.

Fig. 1 shows a typical grid-voltage/grid-current curve ( $AT$ ) for a cold-cathode gas-filled triode. The point ( $v_t$ ,  $i_t$ ) at which the valve fires is indicated by  $T$ . The

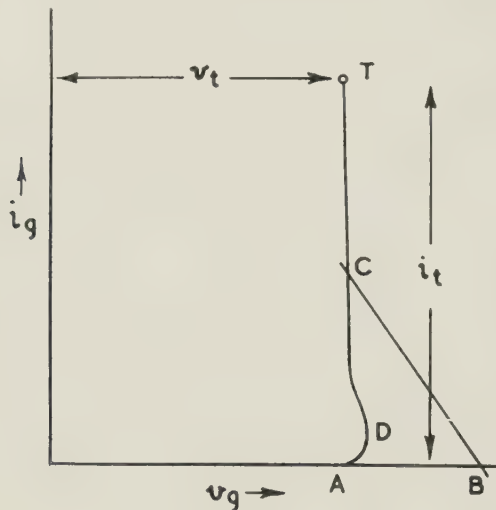


Fig. 1. Grid-characteristic for cold-cathode gas-filled valve

grid current,  $i_t$ , at this instant is known as the transfer current. The line  $BC$  is the load line for the resistance ( $R_s$ ) in the grid circuit (see Fig. 2). If an input voltage  $v_s$  is applied to the grid through  $R_s$ , the grid voltage  $v_g$  is given by  $(v_s - i_g \cdot R_s)$  where  $i_g$  is the grid current. Hence, for the tube to fire,  $i_g \geq i_t = (v_s - v_t)/R_s$  and, if  $R_s$  is large,  $v_s$  must be large. In the example given in Fig. 1, as  $v_s$  is increased the intersection of the two

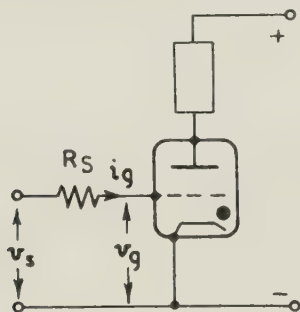


Fig. 2. Gas-filled triode with high grid-resistance

curves moves steadily towards the point  $T$ , but, if the slope of the line  $BC$  is greater than that of the line joining  $TD$ , the tube will fire as soon as  $D$  is reached, because the grid current will snap from  $D$  to a value greater than  $i_t$ .

To take a practical example, consider the photocell circuit shown in Fig. 3. For a vacuum photocell, the anode current depends mainly on the intensity of illumination and only slightly on the anode voltage. Hence, for low illumination levels and normal anode voltage,

the photocurrent will probably be insufficient to supply the transfer current. In these circumstances the sensitivity of the circuit will be limited by the grid characteristics of the valve and not by the value of  $R_L$ . Assuming that the resistance  $R_L$  is much greater than that of the grid-cathode path of the valve, the minimum

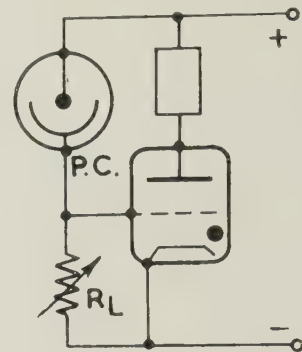


Fig. 3. Typical application with photocell

illumination which will fire the valve is that which produces a photocurrent slightly greater than the transfer current. As the latter is generally high (10 to 100  $\mu A$ ), the circuit will not be very sensitive.

The limitation imposed by the transfer current can be greatly reduced by the application of a train of short voltage pulses to the grid of the valve from a low impedance source as shown in Fig. 4. Here the level of the pulses is determined by the input voltage  $v_s$  and the transfer current is provided by the pulse source. If

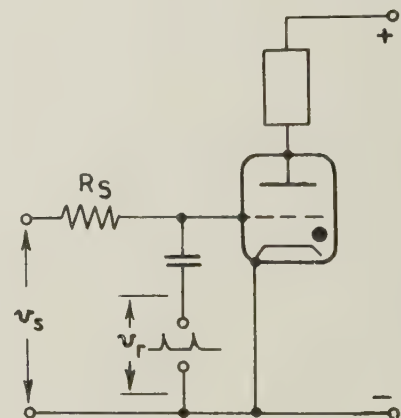


Fig. 4. Circuit of Fig. 2 with the addition of pulse voltages

the amplitude of the ripple voltage  $v_r$  is about 10 V peak, grid current will flow only during the peaks of the pulses and, by making the duration of the pulses short compared with the interval between them, the average grid current can be reduced substantially. By this means, the maximum value of  $R_s$  which can be usefully employed may be increased a hundredfold. In practice, a convenient source of ripple is available in the unsmoothed output from the rectifier supplying the h.t. to the circuit. By differentiation, a train of sharp pulses,



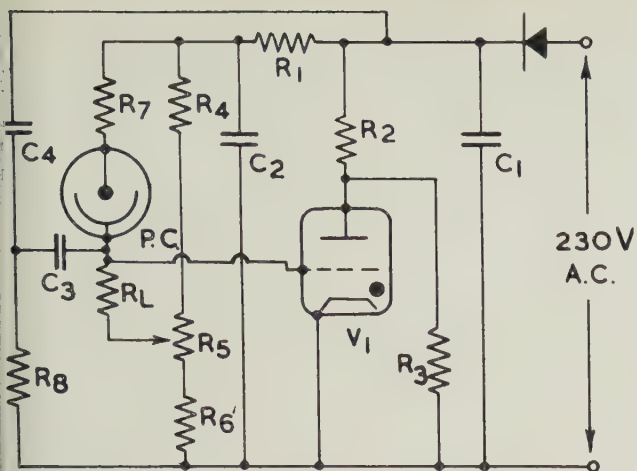


Fig. 5. Photoelectric relay in which the pulses are derived from the rectifier ripple voltage

$R_1 = R_3 = 68 \text{ K}\Omega$	$R_2 = 22 \text{ K}\Omega$	$R_4 = R_6 = 15 \text{ K}\Omega$
$R_5 = 20 \text{ K}\Omega$	$R_7 = 1 \text{ M}\Omega$	$R_8 = 100 \text{ K}\Omega$
$R_L = 1\text{--}120 \text{ M}\Omega$	$C_1 = 1 \mu\text{F}$	$C_2 = 8 \mu\text{F}$
$C_3 = 0.001 \mu\text{F}$	$C_4 = 0.002 \mu\text{F}$	$V_1 = \text{G240/2D}$
P.C. = CMG8	Rectifier = H.75.	

0 or 100 per sec., is obtained. Fig. 5 shows a circuit in which the pulses are derived in this way. By adjusting

the tapping of  $R_5$  to bias the grid to a potential just less than  $(v_t - v_r)$ , the relay valve can be made to fire when a light of very low intensity falls on the photocell.

The principle of "pulsing" has also been used to advantage in the design of a liquid level indicator. In this case, the current flowing through probe electrodes in the liquid had to be kept low to minimize electrolytic effects. A high resistance was therefore connected between the valve grid and one probe, the other probe being taken to a positive supply. When the liquid level rises to touch both probes, the grid is biased positively and the pulses provide the transfer current to trigger the valve.

It is important to remember that the value of  $v_s$  which will trigger the valve depends on the value of  $v_r$ . If the pulses are derived from the rectifier as in Fig. 5, the value of  $v_r$  will vary with the supply voltage. This effect has been applied to the circuit of a process timer, the time interval of which was required to depend on the supply voltage in a particular manner. It is hoped to publish a full account of this application shortly.

#### ACKNOWLEDGMENT

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## NOTES AND NEWS

### New Books

**The Physics of Rubber Elasticity.** By L. R. G. TRELOAR, Ph.D., F.Inst.P. (London: Oxford University Press.) Pp. 254. Price 21s. net.

The rapid expansion of the theory of rubber elasticity, since the kinetic theory was put forward by Meyer, von Susich, and Valko, has added to physics a body of knowledge which should now take its place side by side with the kinetic theory of gases and the theories of Einstein, Debye, Born and Blackman for crystalline solids. The author of the present volume has himself, as a member of staff of one of the most active centres of research on rubber, the British Rubber Producers' Research Association, made important contributions to the application of the kinetic theory.

The purpose of the book is to give the kinetic theory of rubber elasticity, in all its aspects, in a form which will be understood by the average industrial physicist and by his academic colleague who is not a specialist in this field. This purpose is admirably fulfilled, the physical aspects of the theories being presented with great clarity. The lengthy and complex mathematical studies associated with the statistical treatment of rubber elasticity are very wisely omitted. Once the physical basis of any aspect of the theory has been formulated, the author proceeds straight to the result of the purely mathematical argument. This method of presentation enables the physical connexions between assumptions and the results to be understood and focuses attention on the weak and strong points of the theories.

After an introductory chapter outlining briefly the historical background and the basic phenomena of rubber elasticity,

the thermodynamic relations are deduced, which enable the entropy and internal energy changes during deformation to be calculated from experimental measurements. The results show the predominant role of the entropy term in the free energy and hence in the stress produced in any deformation. The elastic properties of long chain molecules are then deduced using the concept of the random chain, in which the angle between successive links can take any value. The corrections needed to bring this theory nearer to the behaviour of a real molecule are discussed and then the theory of a three-dimensional network is given in a form suitable for deriving the stress corresponding to the most general three-dimensional homogeneous strain. At this stage the concept of the "stored energy function," which equals the reversible work of deformation, is introduced. The experimental evidence is then shown to be compatible with the simple statistical theory for comparatively small strains only and modifications to the theory are introduced. A more complex stored-energy function, based on an equation of Mooney, gives a better representation of the results of various experiments for large strains, but this modification has not yet been related to the statistical theory. The measurement of strain-birefringence is shown to be a valuable technique, in which the birefringence can be related quantitatively to stress and strain and also used to obtain information on the flexibility of the molecules. The remaining chapters outline experimental work on the crystallization of rubber and its relation to mechanical properties, stress relaxation, flow and dynamic properties. The quantitative interpretation of these experimental results from the properties of the rubber molecule has

not been very successful, but physical ideas have been developed to explain the qualitative results. The last chapter gives rather more fully Rivlin's theory of large elastic strains in terms of the stored-energy function.

To condense into some 250 pages a clear and stimulating account of so wide a field of contemporary physics is an achievement. The book will be of value to physicists, but it will also be intelligible to the physiologist, biologist and biochemist whose interest is in the properties of living tissues. By making accessible the background ideas of the theory of rubber elasticity, it is probable that applications of the theory will be made to the many rubberlike materials. The interfering effects of changes of internal energy for some such systems will, however, impose a need for some caution in this respect. As is natural in a book on rubber elasticity, this aspect has not been greatly emphasized.

ALAN G. WARD

**Foundations of Modern Physics** (2nd ed.). By T. B. BROWN, Ph.D. (New York: John Wiley and Sons, Inc.; London: Chapman and Hall, Ltd.) Pp. xvi + 391. Price 40s. net.

This book is based on "the fourth unit of a two-year general course in physics" at the George Washington University. The basis throughout is the description of the original experiments on which the advances were based and sufficient "classical" material is included to make the book self-contained. Only elementary algebra and geometry are required to follow the mathematics and to work the many exercises.

The text, which has been brought up to date (1948) in this

revision, is well written and covers a wide field, including, for example, kinetic theory and polarized light. It can be recommended to the non-specialist requiring a wide, but not too deep, account of the present state of physics but is perhaps too expensive for students in this country. H. R. LANG

**An Introduction to Molecular Spectra.** By R. C. JOHNSON, M.A., D.Sc. (London: Methuen and Co. Ltd.) Pp. xiii + 296. Price 40s. net.

Dr. Johnson's well-known monograph on "Atomic Spectra" is now followed by a much larger and very welcome book on the more complex subject to which he contributed many experimental investigations. It provides graduate students in physics or chemistry with a sound basis for further study or practical work within the limits of an ordinary mathematical equipment. Although results of wave mechanics are introduced at a few points, familiarity in the use of the method is not assumed and in the treatment of electronic structures of polyatomic molecules matters involving a knowledge of Group Theory are excluded.

The first twelve chapters deal systematically and thoroughly with the structures and band spectra of diatomic molecules; they are followed by one very long chapter on polyatomic molecules and their spectra. The remaining two chapters deal with Raman spectra and with various applications of molecular spectroscopy in astrophysics, chemistry and bio-chemistry.

The text contains 150 line diagrams and is further illustrated with eight plates of well chosen and excellently reproduced spectrograms. The selection of material, the manner of presentation, and the printing and production are all admirable. W. J.

## Journal of Scientific Instruments

### Contents of the February Issue

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- A Stabilizer for Proportional Counters. By D. H. Wilkinson.
- An Electrical Transient Display System. By J. A. Lyddiard and J. W. Osselton.
- An Electrical Viscometer. By A. R. Boyle.
- An Electronic Stimulator for Biological Research. By V. H. Attree.
- Platinum Resistance Thermometers of Small Dimensions. By C. R. Barber.
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- Double-Trace Scanning Microphotometer for Precision Measurement of Line Position. By T. B. Rymer and J. S. Halliday.

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## British Journal of Applied Physics

### Original Contributions accepted for publication in future issues of this Journal

- Solution of Partial Differential Equations with a Resistance Network Analogue. By G. Liebmann.
- Note on the Rheological Properties of Elasto-Plastic Materials. By K. Goldsmith.
- Measuring a Large Displacement by Interferometry. By H. Barrell and M. J. Puttock.
- The Formation and Crystal Structure of Silicon Carbide. By A. Taylor and D. S. Laidler.

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## Summarized Proceedings of a Meeting on "The Investigation of Biological Systems by the Electron Microscope and by X-ray Analysis"—Buxton, 1949

During The Institute of Physics' convention held in Buxton in May 1949, the Electron Microscopy and X-ray Analysis Groups held a joint session which was devoted to a discussion on "The Investigation of Biological Systems." The papers and the discussion on them are summarized in this report.

In his introductory survey, PROFESSOR J. T. RANDALL (King's College, London) showed the usefulness of the manifold approach to complex problems in biophysics. Commenting on X-ray diffraction methods as applied to solutions of proteins, colloids and other systems of biological interest, the speaker briefly mentioned Kreger's recent micro-diffraction technique<sup>(1)</sup> and then passed on to a discussion of low-angle scattering from protein solutions and colloidal suspensions. This method could provide information on the size and shape of molecules as well as statistical data about their distribution. Since the particles concerned were, in general, large compared to the wavelength of X-rays used, destructive interference caused the intensity of scattered X-rays to fall off steeply with angle and the diffraction effects had, therefore, to be sought at very small angles. If the particles were sufficiently far apart and arranged at random, the type of diffraction curve obtained resembled that of a gas, since the scattering at angles near to zero was the sum of the scattering by individual particles.

At high particle-concentrations a short-range order of particle distribution might arise and produce diffraction effects similar to those produced by simple liquids (such as liquid metals) at larger angles of diffraction. Prof. Randall showed striking diffraction photographs of concentrated solutions of haemoglobin and egg albumin taken by Riley and Herbert<sup>(2)</sup> and also mentioned Riley and Schulman's study of oil-water disperse systems<sup>(3)</sup> as examples of this "liquid" type of scattering.

The speaker criticized the current tendency to make deductions from the "positions" of diffraction peaks by using what is euphemistically called a modified Bragg Law and stressed that the only method which strictly applied was the deduction of the radial distribution function of scattering matter from the general Zernike-Prins expression<sup>(4)</sup> using the method of Fourier inversion. The application of the low-angle scattering method to living cells was limited to those types of cell where one kind of molecule or structure predominated.

As an example of the extent to which combined techniques had been used in the study of a structural problem Professor Randall mentioned work by McEwen and Mould on the properties of gels of bentonite plates. Methods had been devised to measure the rigidity of such a colloid system. The results could only be interpreted with additional information supplied from studies of birefringence, X-ray scattering, optical scattering and electron microscopical structure.

The speaker mentioned Caspersson's work<sup>(5)</sup> on the ultra-violet adsorption spectra of purines and pyri-

midines in nucleoproteins and predicted the extension of this technique, so as to make the rapid survey of ultra-violet<sup>(6)</sup> and infra-red spectra a routine procedure in biophysical investigations of structure. In this connexion Seeds and Wilkins<sup>(7)</sup> had developed a simplified reflecting microscope the objective of which was equivalent to an ordinary  $\frac{1}{8}$  in objective and which had a working distance of 9 mm. With this long working distance, micro-manipulation of biological material was possible.

Some electron micrographs illustrating details in the structure of cell nuclear membranes, spermatozoa and tissue culture cells were discussed.

### THE ELECTRON MICROSCOPY OF VIRUSES

DR. I. M. DAWSON (National Institute for Medical Research, London), in dealing with this subject, said that in the field of virus research the electron microscope had already made a valuable contribution to the study of size and structure. Statistical values of sizes obtained from filtration and centrifugation experiments had been confirmed and direct observation of individual virus particles had shown that the larger animal viruses were complex organisms akin to bacteria whereas the smaller plant viruses had molecular characteristics.

In studying the larger virus particles electron microscopical technique had been somewhat handicapped by comparison with its optical counterpart since few staining methods had been developed for electron microscopy. The use of inorganic heavy metal cations had already proved valuable in studies of fibrous protein structure;<sup>(8,9)</sup> enzymatic digestion had been used to throw into relief certain components in virus structure<sup>(10)</sup> and micro-incineration had been applied to a study of the inorganic constituents of fibrous proteins.<sup>(11)</sup> There was, however, considerable scope for further developments in this field.

For studying the growth and development of virus particles techniques were now being developed to minimize the difficulties inherent in examining dead and vacuum-dried material. Since it was impossible to observe a single virus particle during the whole course of its life-cycle, statistical studies had to be made of large numbers of virus particles at various stages in the developmental cycle. Viruses have been photographed within tissue culture cells<sup>(12,13)</sup> and adsorbed on to the surface of red blood cells.<sup>(14,15)</sup> In studies on tobacco mosaic virus, Williams and Steere<sup>(16)</sup> had refined the technique to minimize the distortion of virus morphology during the specimen preparation. In all these methods the aim was to minimize distortion of virus material in centrifugation and prevent the formation of aggregates

of virus material. Aggregation difficulties were, of course, serious in any statistical study of virus multiplication from particle size measurements.

As results come forward, it was increasingly apparent that there were very few general constants in virus structure. Although viruses might be grouped into general classes with regard to growth and morphology, as, for example, the pox group comprising vaccinia, variola, ectromelia and fowl pox, yet the differences were considerably greater between the larger animal viruses and the smaller plant viruses than between either the larger animal viruses and bacteria or the crystalline proteins and the smaller plant viruses.

#### PROTEINS AND THE ELECTRON MICROSCOPE

DR. R. REED (University of Leeds) surveyed applications of the electron microscope to research on protein structure. The electron microscope could be used to determine the size and shape of protein molecules, thus providing a direct check of the results obtained by X-ray diffraction and other physico-chemical techniques. Unfortunately, the limit of resolution obtainable in practice was only of the order of 50 Å. This meant that, for the present, electron microscopists had to be content with the study of the larger type of protein molecules. Among these, the virus proteins offered one of the most fruitful fields of research, even though information on intra-molecular structure had been disappointingly meagre. The speaker showed examples of R. W. G. Wyckoff's<sup>(17)</sup> spectacular photographs of faces of plant virus crystals and he, as well as other speakers, dwelt on the interesting correlation between X-ray diffraction work and Wyckoff's electron micrographs, which directly confirmed some of the crystallographers' ideas regarding the packing of globular protein molecules in a crystal lattice.

By revealing a relationship between globular and fibrous proteins, electron microscopy and low-angle X-ray diffraction studies had given complementary results of great value. After mentioning some examples, the speaker suggested that many, and possibly all, fibrous proteins might be formed by the aggregation of globular units.

#### RECENT WORK ON STRUCTURE OF PROTEINS

DR. M. F. PERUTZ (Cavendish Laboratory, Cambridge) developed a theme already mentioned by Dr. Reed: the relationship between the fibrous and the globular proteins. The great majority of protein fibres produced by the living cell gave the  $\alpha$ -keratin pattern which seemed to correspond to polypeptide chains of roughly cylindrical shape spaced about 10 Å apart, with a pattern repeating at 5 Å intervals along the chain direction. This pattern probably contained three amino-acid residues.

Electron microscopy had shown that natural protein fibres did not have a micellar structure, as was commonly believed at one time, but that they contained at least one level of organization between the polypeptide chain and the macroscopic fibre. This level consisted of microfibrils whose thickness varied between 100 and 1000 Å in different proteins. These microfibrils, in turn, often appeared to consist of an assembly of

globular particles. It seemed, then, that the cell produced some at least of its protein chains in the  $\alpha$ -keratin configuration. The chains might form more or less globular particles which, in turn, were strung together into microfibrils and these fibrils might finally aggregate into macroscopic fibrils. This was a working hypothesis rather than a statement of fact but it was significant, perhaps, that Kendrew and the speaker had found two typical corpuscular proteins, myoglobin and haemoglobin, to consist of bundles of polypeptide chains folded in what seems to be the  $\alpha$ -keratin structure.<sup>(18, 19, 20)</sup>

Dr. Perutz stressed that the nature of the  $\alpha$ -keratin structure was still unknown. It was important to find out how many polypeptide chain models with a repeat pattern of 3 aminoacid residues/5 Å could be built. Kendrew had recently investigated all the possible spirals with three-fold symmetry which gave the right repeat and arrived at the encouraging conclusion that there may be only two possible ones—the Huggins spiral and one other.

Most important observations bearing on the  $\alpha$ -keratin problem had recently been made by Ambrose and Hanby<sup>(21)</sup>, and Ambrose, Elliott and Temple,<sup>(22)</sup> who had investigated the infra-red absorption of fibrous proteins of the  $\alpha$ -keratin type in polarized light and had found a striking dichroism in the NH stretching frequencies known to be associated with hydrogen bonds. Their observations indicated hydrogen bonds linking carboxyl and imino groups to be oriented preferentially parallel to the chain direction, which meant that the folds in the  $\alpha$ -keratin chains must be held together by hydrogen bonds located *within* each chain.

Finally Dr. Perutz described a recent observation of Barer, Jope and himself, on the absorption spectra of single protein crystals in polarized ultraviolet light. Single crystals of haemoglobin and myoglobin were examined using Burch's new reflecting microscope.<sup>(23)</sup> This instrument made it possible to measure the absorption spectrum of a crystal weighing as little as  $10^{-9}$  g. The speaker demonstrated spectra of haemoglobin crystals showing dichroism in both the 400 and 290 m $\mu$  regions, which were known to be associated with the iron containing haem groups and with the indol groups of tryptophane respectively. The dichroism indicated both these flat groups to be oriented at right angles to the length of the polypeptide chains. A similar tryptophane dichroism had been observed by Butenandt and others<sup>(24)</sup> in streaming solutions of tobacco mosaic virus. The sterical configuration of the polypeptide chains might be such that any large flat group was oriented at right angles to the chain direction.

#### DISCUSSION

DR. D. M. HODGKIN (Department of Chemical Crystallography, Oxford) noted that, in general, there was a wide gap between the X-ray and electron microscope fields, but that there was an intermediate region in which the evidence overlapped and in which collaboration seemed most fruitful. She showed X-ray diffraction photographs of dried and wet tobacco necrosis virus crystals of the



same strain as that examined by Wyckoff<sup>(17)</sup> with the electron microscope. This virus had a molecular weight of about 2,000,000. One of the pictures from a dried crystal showed an almost perfect hexagonal array of reflexions, in good agreement with the array of virus particles which Wyckoff had found on shadow-cast replicas of the corresponding crystal face. A photograph of the wet crystal contained a rich intra-molecular pattern extending to spacings of the same order as those found in crystals of other proteins of smaller molecular size.

MRS. HODGKIN also mentioned recent results obtained by Carlisle<sup>(25)</sup> on tomato bushy-stunt virus crystals. These suggested that each virus particle consisted of a number of close-packed sub-units which were not yet visible on electron micrographs. An increase in resolution to distances of the order of 20 Å would reveal much interesting detail in the fine structure of viruses.

DR. PERUTZ, referring to the introductory address by Professor Randall, wondered whether all reflexions except the innermost one on Dr. Riley's low-angle scattering photograph of egg albumin solution were perhaps of intra-molecular origin. He suggested that, in order to distinguish between intra- and inter-molecular reflexions, the change in the diffraction patterns with protein concentration should be investigated.

DR. PERUTZ also wondered whether Dr. Riley had considered an alternative explanation of the low-angle scattering pattern of human haemoglobin. There was reason to believe that this protein molecule was more iso-diametric than horse haemoglobin. Suppose the haemoglobin molecules in Dr. Riley's very concentrated solution were simply cubic close packed and it were impossible to resolve powder lines which were close together. In this case, the combined 111 and 200 reflexions might appear as the first line and the combined 220 and 311 reflexions as the second one. The ratio of the spacings of these two pairs of unresolved lines would be 1.65 : 1, exactly as observed.

DR. C. H. CARLISLE (Birkbeck College, London) mentioned that Patterson projections recently obtained from wet ribonuclease pictures were compatible with an arrangement of polypeptide chains similar to the one described by Dr. Perutz in haemoglobin. Moreover there was evidence that the chains had a spiral configuration of the trigonal type.

DR. H. LIPSON (Manchester College of Technology) wondered why radiation of longer wavelength such as Aluminium K $\alpha$  was never used in the investigation of biological substances with large spacings.

DR. I. MACARTHUR (University of Leeds) replied that he had experimented with aluminium radiation some years ago and had found it to have a disintegrating effect on proteins.

DR. DAWSON referred to the areas on the surface of vaccinia virus<sup>(10)</sup> where the electron microscope had revealed sub-units arranged with a certain regularity. He wondered whether X-ray diffraction had thrown any light on the structure of this virus.

DR. MACARTHUR reported on low-angle scattering of X-rays from vaccinia virus. Using the techniques developed by Guinier and his collaborators, Dr. MacArthur had been able to correlate the surface morphology of the virus with the X-ray pattern. This had shown the particle-size distribution within the virus to be somewhat random.

DR. PERUTZ mentioned that he had been interested whether a regular distribution of sub-units within the virus particle would give rise to a diffraction maximum at low angles, rather like the maximum found in the study of red blood cells. X-ray photographs taken by Mr. Nelson had not revealed the existence of any maximum in the low-angle scattering curve.

Hence the sub-units were not likely to be of uniform size.

I. M. DAWSON, M. F. PERUTZ

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## ORIGINAL CONTRIBUTIONS

## Electrode Erosion by Spark Discharges\*

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Theoretical estimates of the areas of electrode hot-spots are made from a consideration of the lateral spread of the electrons and positive ions due to diffusion and their space charge. Areas thus found show that very high current densities of the order  $10^6$  A/cm<sup>2</sup> are possible in typical short spark gaps at atmospheric pressure. The energy balance at such hot-spots for an equilibrium temperature equal to the boiling point of the electrode material is then considered and, neglecting chemical attack, an equation is set up relating the volume of electrode evaporated per spark (i.e. the spark erosion) to the physical properties of the electrode material.

The production of electrode vapour in arc and spark discharges, and the resulting erosion of the electrodes, have been studied considerably during recent years.<sup>(1, 2, 3, 4)</sup> It is the purpose of this paper to examine the relevant parameters affecting electrode erosion in the simplest case, to propose a theory of the mechanism and to discuss this in the light of available experimental data.

## ELECTRODE CURRENT DENSITY

It is of interest first to make an estimate of the cross-section of the current on the basis of the lateral diffusion of ions and electrons in the spark track. In accordance with the Similarity Theorem, the ratio of the current density  $J$  to the gas pressure  $p$  is constant for any given discharge potential  $V$  in geometrically similar systems, so that  $J$  is low at low pressures, giving rise to the well-known glow; at high pressures  $J$  can be high, giving rise to the well-known thin track. Since most of the practical applications of spark discharges occur at pressures greater than or equal to atmospheric, only such pressures will be considered here. Further, if the gap is short it is reasonable to assume a comparatively simple mechanism of sparking, viz. the progressive development of an electron avalanche and the motion of the simultaneously produced positive ions created there back to the cathode.

Consider, for simplicity, a point source of electrons on a cathode of a parallel-plate gap of length  $d$ . The mean energy of agitation  $eE_1 (=mu^2/2)$  is determined by the ratio of field strength  $Z (=V/s)$  to the gas pressure  $p$ . The order of magnitude of the lateral spread of the avalanche may be obtained by considering the diffusion of a number of electrons, released at a point, as they travel along the axial electric force  $Z$ .<sup>(5, 6, 7, 8, 9)</sup> If  $n$  is the electron concentration, and  $\alpha$  the primary ionization coefficient, the equation of motion is

$$\partial n/\partial t = \alpha W_1 n - \operatorname{div} J \quad . \quad . \quad . \quad (1)$$

where  $J$  is the true electron current-density vector to an observer at rest.  $J$  is given by

$$J = -D_1 \operatorname{grad} n + neW_1(Z') \quad (2)$$

where  $Z'$  is the resultant electric field due to the charges on the electrodes, and also to any space charge in the gas,  $D_1$  is the coefficient of electron diffusion and  $W_1(Z')$  is the electron velocity in the direction of the resultant force  $Z'$ . To an observer moving with velocity  $W_1$  along the direction of  $Z$ , the current-density vector will be  $J_1$  given by

$$J_1 = J - neW_1 \quad (3)$$

and for him  $\partial n/\partial t = \alpha W_1 n - \operatorname{div} J_1$ .

Using equation (3) it follows that

$$\begin{aligned} \partial n/\partial t &= \alpha W_1 n - \operatorname{div} (J - neW_1) \\ &= \alpha W_1 n + D\nabla^2 n - \operatorname{div} n[W_1(Z') - W_1(Z)] \quad (4) \end{aligned}$$

In the electron avalanche the space charge may be neglected owing to the presence of positive ions, so that  $Z' = Z$ . It is seen from equation (4) that a quantity  $n = n_0 \exp \alpha W_1 t$  satisfies the equation

$$\partial n_0/\partial t = D\nabla^2 n_0 \quad (5)$$

This is the diffusion equation which the electrons would satisfy if there were no internal generation (by ionization), i.e. if the total number remained  $n_0$ . If, therefore, the spatial distribution of  $n_0$  is calculated at any time  $t$  by solving equation (5), then, since  $n = n_0 \exp (\alpha W_1 t)$ , it is clear that the spatial distribution of  $n$ , the number of electrons including those internally generated, is the same as that of  $n_0$ , i.e. it is also given by equation (5).

Writing  $r$  for the radial distance from the axis, and defining  $R^2$  by the relation  $NR^2 = \int nr^2 d\tau$ , it follows that  $Nd(R^2)/dt = D_1 r^2 \nabla^2 n d\tau$ , which, using Green's theorem gives

$$d(R^2)/dt = 4D_1.$$

The cross-section  $A_1$  of the electron avalanche at the anode is then given by  $A_1 = 4\pi D_1 s/W_1$ .

$D_1 = lu/3$  approximately and  $W_1 = 2eZl/3mu$  approxi-

\* Based on a lecture to the South Wales Branch of the Institute of Physics at Cardiff on 9 February, 1949.



nately, where  $l$  is the mean free path, so that  $D_1/W_1 = mu^2/2Z = dE_1/V$  approximately.

Thus:  $A_1 = 4\pi d^2 E_1/V$  . . . . . (6)

It is now necessary to consider the motion of the positive ions. The high concentration at the anode will move to the cathode where the electron concentration is much lower; consequently, it is not possible to neglect the space charge, which will now affect the motion by increasing the lateral diffusion, so dispersing the cloud. It is necessary to estimate the magnitude of this effect, in order to calculate the area of the cathode struck by the positive ions (which will influence the area through which the positive ion current enters the cathode).

The equation of motion of the positive ions produced in the avalanche is obtained from equation (4) by setting  $v = 0$ , so that  $\partial n/\partial t = D_2 \nabla^2 n - \text{div } n[W_2(Z') - W_2(Z)]$ .

But  $W_2(Z) = \beta Z$ , where  $\beta$  is the positive ion mobility. Put  $Z'' = Z' - Z$ ,  $\partial n/\partial t = D_2 \nabla^2 n - \text{div } \beta n Z''$ , where  $Z''$  is now the field due to the space charge and its multiple images in the electrodes. Now  $\text{div } Z'' = 4\pi ne$ , hence  $\partial n/\partial t = D_2 \nabla^2 n - 4\pi \beta n^2 - \beta(Z'' \text{ grad } n)$ .

To obtain a solution of this equation, consider a

Integration gives  $r_t^2 - r_0^2 = 4e\beta Nt$ , i.e.

$$(A_2)_t - (A_2)_0 = 4\pi e\beta Nt = 4\pi ed^2 \alpha (\exp \alpha d)/2V \quad (8)$$

It is assumed that the avalanche was generated by the emission of a single electron from the cathode, and if  $N$  is given the approximate value  $\alpha(\exp \alpha d)/2$ , and  $(A_2)_0$  the value of  $A_1$  given by equation (6), and set  $t = d^2/\beta V$ , the time for a positive ion to cross the gap, then the area  $(A_2)_t$  given by equation (8) may be regarded as the area of the resulting cathode hot-spot, and  $r_t = \sqrt{[(A_2)_t/\pi]}$  its radius.

In order to fix ideas, some specific examples of practical interest will be considered, namely: short gaps of 0.2 mm and 0.5 mm in air at atmospheric pressure and, to represent an ignition spark plug, a 0.3 mm gap at a pressure of 3 atm., and, for comparison, a 1 cm gap in air at 1 atm., still assuming that the ionization processes are the same. The data of sparking potentials<sup>(11)</sup> ionization coefficient<sup>(5,12)</sup> assumed and the electron avalanche and ion current cross-sections calculated, are given in Table 1.

Thus, in typical short gaps at atmospheric pressure it is seen that the space charge enlarges the first positive

Table 1. Spark data and calculated hot spot areas

$d$ (mm)	$V$ (V)	$E_1$ (eV)	$\alpha$	$A_1$ cm <sup>2</sup> from equation (6)	$(A_2)_t - (A_2)_0$ cm <sup>2</sup> from equation (8)	$r_t$ (cm)
0.2	1 500	6.7	494	$2.2 \times 10^{-5}$	$2.4 \times 10^{-6}$	$2.8 \times 10^{-3}$
0.5	2 700	5.0	175	$5.0 \times 10^{-5}$	$8.4 \times 10^{-7}$	$4.0 \times 10^{-3}$
10	32 000	3.5	17	$1.4 \times 10^{-3}$	$1.0 \times 10^{-2}$	$6.0 \times 10^{-2}$
*0.3	4 800	5.0	525	$1.1 \times 10^{-5}$	$7.0 \times 10^{-4}$	$1.5 \times 10^{-2}$

at  $p = 3 \times 760$

\* Typical ignition spark plug conditions.

column of ions formed in the original avalanche. In typical short gaps, the data of Table 1 show that  $Z/p$  is about 40 to 100 V/cm/mm of mercury, and under these conditions  $E_1$  lies between about 4 and 6 eV<sup>(10)</sup>. Unless the gas is greatly heated, the mean energy of the positive ions  $eE_2$  is unlikely to exceed about 0.1 eV. Consequently, equation (6) shows that the ratio of the cross-section  $A_2$  of a positive ion beam to  $A_1$ , that for electrons under the same conditions, is  $\approx 0.025$ . Hence, the effect of diffusion of the ions may be neglected and any dispersal must be due to the space charge. Again, it also follows that the radius of the area  $A_1$  of the current cross-section is small compared with  $d$ , since  $E_1/V$  is small.

Thus, no great error is made in assuming that (to an observer moving with the ion stream along  $Z$ ) the main components both of the space charge field  $Z_1$  and of the concentration gradient  $\text{grad } n$  are both radial and perpendicular to the axial field  $Z$ . This assumption allows a simplified picture of the ion motion to be made, viz. that of the lateral expansion of a thin column of ions dispersing under its own space charge. Clearly, since diffusion can be neglected, we note that the radius  $r$  of a cylindrical surface enclosing, per unit length, a fixed number  $N$  of ions per cm expands at the rate  $dr/dt = \beta Z_1 = \beta 2eN/r$ .

ion beam only a little to about  $5 \times 10^{-5}$  cm<sup>2</sup> at the cathode and it is from this area that the next burst of electrons is largely drawn, whilst at higher pressures, or for longer gaps, the effect of the space charge can be considerable. A small number of such successive avalanches are sufficient to produce breakdown and, when once established, the active hot area is unlikely to be increased, so that on this view the area of the cathode current cross-section in typical short gaps would be approximately of the order  $5 \times 10^{-5}$  cm<sup>2</sup> and the radius of the hot-spot would be about  $3 \times 10^{-3}$  cm.

In a typical spark plug, for instance, when no series resistor is used, the spark peak current can be of the order 100 A and this involves a cathode current density of the order  $10^6$  A/cm<sup>2</sup>, whilst for lower currents of 1 A the value of  $J$  would be about  $10^4$  A/cm<sup>2</sup>. The area  $(A_2)_t$  given in Table 1 for a 1 cm gap was calculated on the assumption of a continuous avalanche development across the gap, just as with the short gaps. As this process is much less likely in a 1 cm gap, the areas calculated for such a gap must therefore be regarded as outside upper limits.

The above estimates of the width of the cathode spot and of the current density are of the same order as those measured experimentally over a wide range of conditions. Froome<sup>(2)</sup> in an 18.5 mm gap at low pressures has

observed hot-spot diameters of  $5 \times 10^{-3}$  cm in general agreement with those deduced in Table 1, giving current densities from  $10^2$  A/cm<sup>2</sup> to values greater than  $10^6$  A/cm<sup>2</sup>. Cobine and Gallagher<sup>(13)</sup> using low pressures observed hot-spot widths from  $5.4 \times 10^{-6}$  to  $2.75 \times 10^{-2}$  cm which are also of the same order as the values of  $r_t$  given in Table 1 and which involved current densities from  $2 \times 10^4$  A/cm<sup>2</sup>. This general agreement may thus be taken as evidence in support of the broad picture of the spark breakdown factors, discussed above, in which the positive ion current and the electron current bombard a small area of the cathode and anode respectively and it is now necessary to consider the consequences of this.

#### ENERGY DISSIPATION AT ELECTRODES

Chemical reaction, such as oxidation, between the electrode and the gas will be neglected and so will any reaction between the gas atoms and atoms evaporated from the hot-spot. An approximate upper estimate of the time required to raise the hot-spot temperature to the boiling point may be made by considering the rise in temperature of the surface of an infinite metal plane due to a constant heat input. With a material of thermal conductivity  $k$ , specific heat  $s$  and density  $\rho$ , all constants, the temperature rise  $\theta^\circ$  after a time  $t_1$  is given by the equation

$$\theta = 2Q\sqrt{(t_1)/\sqrt{(\pi k s \rho)}} \quad (9)$$

when energy is supplied at the constant rate  $Q$  per unit area, provided that  $\sqrt{(r^2/Kt_1)}$  is large compared with unity,  $K$  being  $k/s\rho$ . Consider a typical metal (nickel) for which  $\theta \simeq 3,000^\circ$ ,  $k s \rho \simeq 1$ , and a spark for which the total current density  $\simeq 10^6$  A/cm<sup>2</sup>. The positive ion current density may be taken as  $10^5$  A/cm<sup>2</sup> and the cathode fall  $\simeq 20$  V, so that at the cathode  $Q \simeq 2 \times 10^6$  W/cm<sup>2</sup>, and at the anode  $\simeq 2 \times 10^7$  W/cm<sup>2</sup>; these values lead to values of  $t_1$  of  $2.5 \times 10^{-6}$  sec at the cathode and  $2.5 \times 10^{-8}$  sec at the anode. Further, both such values make the quantity  $\sqrt{(r^2/Kt)}$  large compared with unity, so that the above equation for  $\theta$  is applicable to the case of a hot-spot of radius  $r$  on a plane conductor. It will be noted that, owing to the greater current and also because the electron avalanche strikes the anode before the positive ion avalanche strikes the cathode, the anode hot-spot will be formed more quickly and sooner than the cathode hot-spot. Since most spark discharges in practical applications will last for times exceeding  $1 \mu\text{sec}$ , and typical times are usually  $10^{-5}$  to  $10^{-4}$  sec, the times necessary to establish the hot-spot may be regarded as negligible in comparison. Such discharges are generally due to the rapid quenching of the arc, or to a lack of sufficient energy from the source of e.m.f. to maintain a prolonged arc. Some form of high-frequency sparks, the discharges in some types of enclosed spark gaps and the ignition spark in internal combustion engines are examples of electrical discharges with very short arc phases, sometimes of comparatively high current.

Observation of spark and arc discharges show that

hot-spots, when once established, last as long as the current is maintained. Consequently, the problem of the energy exchanges at the hot-spot may be simplified as follows: a hot-spot of area  $A_1$  at the temperature of the boiling point of the electrode material is instantaneously established on the electrode; then, for a succeeding time  $t$  which may be as large as  $10^{-3}$  sec, depending upon the particular application, the hot-spot is maintained and evaporation takes place from the area  $A$ . At this temperature an equilibrium can be considered as occurring between the rate of supply of energy from the current and the rate of dissipation by absorption as latent heat of evaporation, radiation from the hot-spot and thermal conduction.

Let  $W$  joules be the total amount of energy released by the spark at the two electrodes in the form of kinetic energy of electrons at the anode and as kinetic and potential energy of positive ions at the cathode. Then

$$W = \frac{1}{2}C_e V^2 \text{ joules} = cV^2 \text{ calories} \quad (10)$$

where  $V$  is the sparking potential of the gap (in V), and  $C_e$  is the effective gap capacity (in farads) discharged by the spark to the electrodes and  $C = C_e/8.4$ .  $C_e$  is not the total capacity in parallel with the gap and is considerably less than that. To avoid the necessity of finding the distribution of the liberated spark energy between the two electrodes, the erosion of both will be considered together; the widening of the gap is the only factor of practical importance.

Consider now the dissipation of the energy which the spark releases at the electrodes. The energy given to a hot-spot can be lost by any or all of the following processes:

- (i) Energy is required to heat up a certain mass of electrode to its boiling point, and further energy is absorbed as latent heat of evaporation of the eroded quantity of material. Let this total amount be  $E$  cal.
- (ii) Energy is radiated from the glowing hot-spot as long as it lasts. Let this occur at the rate  $R$  cal/sec.
- (iii) Energy is conducted away from the hot-spot by metallic thermal conduction along the electrodes while the hot-spot lasts. Let this occur at the rate  $H$  cal/sec.
- (iv) Energy is lost from the hot-spot by convection and conduction in the gas in contact with the hot-spot.
- (v) Energy is absorbed as heat by evaporation of electrons which constitute the net thermionic emission from the cathode; also energy is similarly absorbed by any evaporation of positive ions from the anode, reduced by any compensating processes such as the absorption of electrons.

Consider these processes in the above order. The energy required to evaporate a mg. of electrode material can be expressed by the equation

$$E = ms_1(\theta_1 - T) + mL_2 + ms_2(\theta - \theta_1) + mL_1 \quad (11)$$



where  $T$  = steady temperature in degrees Kelvin of parts of electrode remote from the hot-spot,  
 $\theta$  = boiling point of material in degrees Kelvin,  
 $\theta_1$  = melting point of material in degrees Kelvin,  
 $s_1$  = average specific heat in solid state,  
 $s_2$  = average specific heat in liquid state (if any),  
 $L_2$  = latent heat of fusion (if material is melted),  
 $L_1$  = latent heat of evaporation.

The total radiation of energy from the hot-spots on the electrodes takes place at the rate

$$R = \sigma A(\theta^4 - T^4)/4.2 \times 10^7 \text{ cal/sec} \quad (12)$$

where  $A$  is the area of the hot-spots and  $T$  is the absolute temperature of the surroundings. This temperature can be taken to be the same as that of those parts of the electrode which are maintained approximately at a steady temperature. Thus  $T$  in equation (11) is the same as  $T$  in the equation (12) and  $\theta$  is the hot-spot temperature, i.e. the boiling point, as in equation (12). If the electrodes were black bodies,  $\sigma$  would be Stefan's constant =  $5.7 \times 10^{-5}$  ergs/cm<sup>2</sup>/sec/degree<sup>4</sup>, but the metals usually employed are not perfectly black, so that  $\sigma$  will be less than this value. When the hot-spots in the anode and cathode  $A_1$  and  $A_2$  have not equal area, put  $A = A_1 + A_2$ , and no great error is involved in this simplification.

The loss of energy from the hot-spot will now be considered. If the small area  $A$  of radius  $r$  be suddenly raised from the temperature  $T$  to  $\theta$  and then maintained there, and if the quantity  $\sqrt{(Kt/r^2)}$  is a small fraction, the total heat which has passed into the metal at the time  $t$  is approximately  $2(\theta - T)r^2\sqrt{(\pi s \rho k t)}$ , where  $k$  = thermal conductivity,  $s$  = specific heat,  $\rho$  = density, and  $K = k/s\rho$ . But if  $\sqrt{(r^2/Kt)}$  is a small fraction the total heat at the time  $t$  is approximately  $2\pi r(\theta - T)kt$ . For typical electrode materials,  $K$  is approximately unity, and if  $r$  is taken to be about  $3 \times 10^{-3}$  cm, as in short gaps, then  $\sqrt{(r^2/Kt)}$  is small provided  $t$  exceeds about  $10^{-4}$  sec. If consideration at this stage is limited to discharges for which  $t$  has such values, the process (iii) representing the loss of energy by metallic conduction can be expressed by the relation

$$H = 2\sqrt{(\pi A)}(\theta - T)kt \text{ cal/sec} \quad (13)$$

In process (iv), the coefficient of thermal conductivity of gas is negligible compared with that for the electrode material, so that this process can be ignored. With regard to the factors (v), any net loss of energy at the electrodes due to loss of electrons or ions can be deducted from the total energy released by the spark at the electrodes, consequently these losses can be absorbed in the expression for the net energy liberated, which is then given by  $W$  in equation (10).

It is now necessary to consider the energy balance at the hot-spot.

#### ENERGY BALANCE: THE EROSION EQUATION

Since the time  $t_1$  required for the establishment of the hot-spot is small compared with the time  $t$  for which the

hot-spot is maintained, the dissipation of energy by the three important processes of evaporation, conduction and radiation during the time  $t$ , can, without serious error, be equated to the quantity of energy  $W$  which represents the net amount of energy given to the electrodes during the total time  $t_1 + t$ . Thus it follows that

$$E = W - Rt - Ht \quad (14)$$

where  $W$ ,  $E$ ,  $R$  and  $H$  are given by equations (10), (11), (12) and (13). This investigation will be confined to the relation between the total erosion and the total amount of energy released by the spark at both electrodes, assumed of the same material.

Consider the quantity of energy required to heat up and evaporate a mg. of metal from the initial temperature  $T^\circ$  K as given by equation (11). All the factors in this equation are not known accurately, so that simplifying assumptions must be made. For instance, let  $s_1 = s_2 = s$  so that  $s_2$  and  $\theta_1$  can be eliminated;  $s$  is then the average value of the specific heat in the metallic state over the whole temperature range from  $T$  up to  $\theta$ . If the electrode material sublimes, no error is introduced in this way. In any case, for common metals and using Trouton's Rule,  $ms(\theta - T)$  which gives the energy required to heat a mg. up to  $\theta$  is only about one-third of the value of the term  $mL_1$  which gives the energy absorbed as latent heat at the temperature  $\theta$ .

Now, for most metals the value of  $L_2$  is of the order of  $L_1/10$ , and so it may be neglected in comparison with  $L_1$ . The exact values of  $L_1$  are not known accurately for the metals commonly used for electrodes, but it can be eliminated by using Trouton's Rule. This states that for all substances the ratio

$$\frac{\text{latent heat of evaporation of one gram molecule}}{\text{boiling point in degrees Kelvin}}$$

$$= 21 \text{ approximately.}$$

Hence  $ML_1/\theta = 21$ , where  $M$  is the molecular weight of the electrode material, which is the atomic weight for non-associating atoms. Hence equation (11) can be written

$$E = ms(\theta - T) + m21\theta/M \quad (15)$$

The rate of loss  $R$  of energy from the hot-spot due to radiation is given by equation (12), so that if the hot-spot lasts for a time  $t$ , then the loss of energy is  $Rt$ , where

$$Rt = \sigma At(\theta^4 - T^4)/4.2 \times 10^7 \text{ cal.}$$

Now  $T^4$  can be neglected in comparison with  $\theta^4$ , since  $\theta \simeq 3000^\circ$  K and  $T \simeq 1000^\circ$  K for the metals commonly used in spark plugs, and may be less for those in spark gaps.

Hence, the radiation term can be given as

$$Rt = b\theta^4 \quad (16)$$

where  $b = 2.38 \times 10^{-8} A\sigma t$ ,  $A$  = total area  $A_1 + A_2$  of the hot-spots on the electrodes and  $\sigma$  = constant depending on the blackness of the metal.

The factor  $\sigma$  will not vary considerably for the various metals commonly used for electrodes, so that for any given sparking conditions, the factor  $b$  can be taken as a constant which is practically independent of the electrode material and of its rate of erosion.

The heat lost by metallic conduction during the total time  $t$  is practically given by the equation (13), and the factors  $2\sqrt{(\pi A)}$ , and  $t$  can be absorbed into one factor,  $g$ , which for the purpose of this investigation can, without serious error, be taken to be independent of the electrode material. So that

$$Ht = gk(\theta - T) \quad (17)$$

The energy balance at the hot-spot which occurs during the time  $t$  for which the hot-spot exists may now be considered. Using equations (10), (14), (15), (16) and (17) it follows that

$$m[(\theta - T)s + 21\theta/M] = cV^2 - b\theta^4 - gk(\theta - T),$$

but  $m = \rho v$ , where  $v$  = volume of material in  $\text{cm}^3$  eroded per spark. Hence,

$$v = \frac{cV^2 - b\theta^4 - gk(\theta - T)}{\rho[(\theta - T)s + 21\theta/M]} \quad (18)$$

where  $b = 2.4 \times 10^{-8} A \sigma t$ ,  $g = 2\sqrt{(\pi A)}t$ , and  $c = c_e/8.4$  is of the order of, but smaller than, the local parallel capacity of the gap, which factors can be taken as independent of the electrode material. Thus, for a given type of spark in identical gaps, equation (18) gives the relative volumes of eroded metal per spark in terms of the physical constants of the electrode materials.<sup>(1)</sup>

#### EXPERIMENTAL DATA

If  $c$ ,  $b$ , and  $g$  were known accurately it would be possible to calculate the mass eroded per spark for any metal, when arcing and chemical reaction are negligible: but such calculations are not at present possible since the time  $t$  and area  $A$  are not known sufficiently accurately. It is thus of interest to find  $c$ ,  $b$ , and  $g$  empirically from three simultaneous equations involving experimental data, for three materials of known, preferably widely differing, physical properties. Such equations can be obtained from the results of Debenham and associates<sup>(15)</sup> on the erosion of spark plug electrodes in internal combustion aero engines and relevant data for suitable materials are given in Table 2, together with the average values of their physical constants assumed for the range  $(\theta - T)$ .

Table 2. Spark erosion data

Material	Mass (= $\rho v$ ) eroded per spark, in $10^{-9}$ gm	$V$ in kV	$\theta^\circ \text{K}$	$k\text{cgs}$	
Platinum	3.23	4.6	4925	0.15	0.05
Nickel	6.65	4.4	3606	0.125	0.17
Iron	11.8	5.7	3778	0.08	0.22
(Austenitic steel)					

Using these data the following values are obtained for the constants in equation (18) above:

$$\left. \begin{aligned} c &= 0.95 \cdot 10^{-12} \text{ F} \\ b &= 2.00 \cdot 10^{-20} \text{ cal}/(^{\circ}\text{K})^4 \\ g &= 0.95 \cdot 10^{-8} \text{ cm. sec} \end{aligned} \right\} \quad (19)$$

It follows that  $C_e (= 8.4c)$  is  $8 \mu\text{F}$  and since the self-capacity of the plug discharged by the spark was  $70 \mu\text{F}$ , this indicates that only 11% of the total spark energy was released at the two electrodes, the remainder being dissipated in gaseous collisions—a result which can be regarded as reasonable.

The relative erosion of nickel and tungsten was also given by Debenham for respective sparking potentials of 3.85 and 5.6 kV and, although tungsten undergoes erosion due to the formation of oxides and their removal by sparks in air, nevertheless it is of interest to calculate the erosion from equations (18) and (19), and to compare the value thus obtained with that measured. Taking for tungsten  $\theta = 5496^\circ \text{K}$ ,  $k = 0.2$ ,  $s = 0.05$ , the calculated volume erosion relative to nickel = 0.27, as compared with the value 0.75 observed: a result which, taking into account the chemical effects, is also not unreasonable. However, the ratio of the sparking potentials was 1.4, so that in these spark plug experiments twice as much spark energy was released with the tungsten electrodes as with the nickel. Hence it may be concluded that, in inert atmospheres and in gaps triggered so as to spark always at the same potential, the relative erosion of tungsten would be much lower—certainly less than 0.27/2 that of nickel—a result which is in accord with observation with enclosed gaps. The relevance of equation (18) to electrical contact erosion has been discussed elsewhere.<sup>(14)</sup>

#### CONCLUSIONS

Each spark breakdown can release an amount of energy at the small localized areas on the electrodes where the spark strikes sufficiently rapidly to produce evaporation of electrode material in quantities of the same order as those observed. The equilibrium temperature at which the important energetic processes occur at the electrode is the boiling point of the electrode material and this is not appreciably affected by variations in the average temperature of the electrode provided that this, for most metals, is less than about  $750^\circ \text{C}$ . When chemical effects are negligible, spark erosion, i.e. production of electrode vapour, is chiefly a thermal process.

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## The Magnetic Field inside a Solenoid

By J. R. BARKER, B.Sc., A.Inst.P., Queen Mary College, London

[Paper first received 22 August, 1949, and in final form 23 September, 1949]

This paper, resulting from a critical examination of past work on solenoids, together with some original work, gives what are believed to be the most convenient formulae for computing the magnitude of the region of uniform field inside the coil. Tables are given with the object of simplifying the task of designing a solenoid.

Iron-free solenoids find considerable application in experimental physics for producing regions of uniform field which are long in the direction of the lines of force, and also for producing extremely large fields (of the order of 100,000 oersteds) over volumes of a few cubic centimetres. This paper gives the most useful formulae together with numerical data for calculations of the field strength and uniformity. References to a few important papers and books are given below.<sup>(1-5)</sup> All the formulae are in the notation of the author's paper on producing uniform fields with several coaxial coils of fairly small winding cross-section;<sup>(6)</sup> comparison of those fields with that of the solenoid is therefore readily possible.

### FORMULAE

The magnetic field due to a system of pairs of coils of vanishingly small winding cross-section, situated co-axially and symmetrically with respect to the origin of co-ordinates is given by

$$H = 2\pi C_1 [1 + (C_3/C_1)(x^2 - \frac{1}{2}y^2) + (C_5/C_1)(x^4 - 3x^2y^2 + \frac{3}{8}y^4) + \dots] \quad (1)$$

$$h = -2\pi xy C_1 [(C_3/C_1) + \frac{1}{2}(C_5/C_1)(4x^2 - 3y^2) + \dots] \quad (2)$$

where  $H$  and  $h$  are the  $x$ - and  $y$ -components of the field strength at the point  $S(x, y)$  of the figure and

$$C_n = \frac{2\sum_j i_j \sin^2 \theta_j \cdot r_j^{-n} \cdot P'_n(\cos \theta_j)}{= 0} \quad \left. \begin{array}{l} \text{for } n \text{ odd} \\ \text{for } n \text{ even} \end{array} \right\} \quad (3)$$

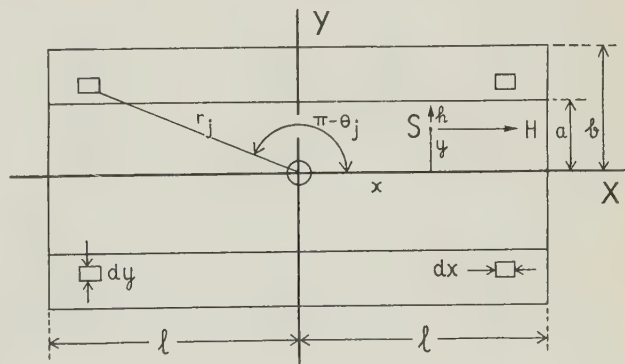
$i_j$  is the current in abamperes (1 abamp. = 10 amp.) in either coil of the  $j$ th pair,  $(r_j, \theta_j)$  and  $(r_j, \pi - \theta_j)$  are the polar co-ordinates of the peripheries of the coils of the  $j$ th pair and  $P'_n(\cos \theta_j)$  is the derivative of the Legendre polynomial of order  $n$  with respect to  $\cos \theta_j$ . Equations 1 to 3 can be deduced with the help of Maxwell's Treatise.<sup>(7)</sup> Equations (1) and (2) hold also for the

solenoid and the  $C_n$  are obtained by assuming the current per unit area of winding cross-section to be uniform, and replacing the summation in equation (3) by an integration. The results, for a thin solenoid of radius  $a$  and length  $2l$  which carries  $i$  abamperes in  $n$  turns/cm., are

$$\left. \begin{array}{l} C_1 = \frac{2nil}{\sqrt{(a^2 + l^2)}} \\ C_3 = -\frac{3nila^2}{(a^2 + l^2)^{5/2}} \\ C_5 = -\frac{5nila^2(4l^2 - 3a^2)}{4(a^2 + l^2)^{9/2}} \end{array} \right\} \quad \dots \quad (4)$$

For a thick solenoid, as in the figure, carrying  $I$  abampere-turns per sq. cm., one finds

$$\begin{aligned} C_1 &= 2Il [\log_e (\sqrt{(b^2 + l^2)} + b) - \log_e (\sqrt{(a^2 + l^2)} + a)] \\ C_3 &= - (I/l) [b^3(b^2 + l^2)^{-3/2} - a^3(a^2 + l^2)^{-3/2}] \\ C_5 &= - (I/12l^3) [b^3(2b^4 + 7b^2l^2 + 20l^4)(b^2 + l^2)^{-7/2} - a^3(2a^4 + 7a^2l^2 + 20l^4)(a^2 + l^2)^{-7/2}] \end{aligned} \quad (5)$$



Thick solenoid showing an elementary pair of turns

In terms of  $i$  and  $n$ , as for the thin solenoid,  $I$  in equation (5) becomes

$$I = ni/(b - a) \quad (6)$$

$C_3$  and  $C_5$  in equation 5 have a far simpler form than that previously published.<sup>(1)</sup>

$C_3$  and  $C_5$  are both much less than  $C_1$  in a long solenoid so that, from equations (1) and (2), a region of uniform field equal to  $2\pi C_1$  exists in the centre of the solenoid.  $h$  need rarely be computed for it has the same order of magnitude as the deviation of  $H$  from  $2\pi C_1$  and produces only a small effect on the direction of the resultant of  $H$  and  $h$  and practically none on its magnitude.

The field at the point  $(c, y)$  which is inside the solenoid but remote from the origin is not given by equations (1) and (2) unless  $\sqrt{c^2 + y^2} < a$ . An expansion can be developed about the point  $(c, 0)$  which is convergent when  $y < a$  whatever  $c$  may be, and it is

$$H = \pi[(C_{1,l+c} + C_{1,l-c}) - \frac{1}{2}y^2(C_{3,l+c} + C_{3,l-c}) + \frac{3}{8}y^4(C_{5,l+c} + C_{5,l-c}) + \dots] \quad (7)$$

In this, the  $C_n$  are taken from formulae (4) or (5), accord-

ing as the solenoid is thin or thick, and the second suffix gives the value to be assigned to  $l$  in those formulae. Dwight's equation 20<sup>(2)</sup> corresponds to equation (7) above for a thick solenoid, but the correlation given here between equations (1) and (7) is new and enables one set of Tables to cover both cases. Gray's equation 28<sup>(4)</sup> corresponds to equation (7) above for a thin solenoid, but the definitions of his  $r_1$  and  $r_2$  should have been interchanged and his  $y^4$ -terms multiplied by  $a$ . As the uniform region almost fills the cross-section of a long solenoid, equation (7) may be simplified by putting  $y = 0$  in practical calculations [e.g., for a thin solenoid with  $a = 1$  and  $l = 7$ , and referring to an origin at the centre of the coil, the field at the point  $(3, 0)$  is  $7\frac{1}{2}$  parts in 1000 less than it is at the centre. At the point  $(3, 0.95)$ , the field is  $1\frac{1}{4}$  parts in 1000 more than it is at  $(3, 0)$ ].

For a thin solenoid only, equation (7) reduces to the well-known exact formula for the field on the axis

$$H = 2\pi ni(\cos \theta_B - \cos \theta_A) \quad (8)$$

where  $\theta_A$  and  $\theta_B$  are the angular polar co-ordinates of the peripheries of the end turns.

#### Set of tables to simplify designing solenoids

$b = 1$				$b = 2$			
	$C_1$	$-C_3/C_1$	$-C_5/C_1$		$C_1$	$-C_3/C_1$	$-C_5/C_1$
3	1.89737	0.01500	0.00206	1.78618	0.02595	0.00240	
4	1.94028	0.005190	0.000456	1.86996	0.01005	0.000692	
5	1.96116	0.002219	0.000133	1.91345	0.004566	0.000232	
6	1.97279	0.001096	0.0000470	1.93855	0.002337	0.0000891	
7	1.97990	0.0006000	0.0000193	1.95423	0.001309	0.0000386	
8	1.98456	0.0003550	0.00000886	1.96464	0.0007863	0.0000183	
9	1.98777	0.0002231	0.00000444	1.97188	0.0004993	0.00000941	
10	1.99007	0.0001470	0.00000238	1.97711	0.0003317	0.00000515	
11	1.99179	0.0001008	0.00000136	1.98103	0.0002286	0.00000297	
12				1.98402	0.0001626	0.00000179	
13				1.98635	0.0001187	0.00000112	
14							
15							
16							

$b = 3$				$b = 4$			
	$C_1$	$-C_3/C_1$	$-C_5/C_1$		$C_1$	$-C_3/C_1$	$-C_5/C_1$
3	1.66177	0.03229	0.00189	1.54232	0.03461	0.00136	
4	1.78272	0.01414	0.000700	1.69042	0.01673	0.000592	
5	1.85067	0.006951	0.000277	1.77993	0.008848	0.000269	
6	1.89184	0.003744	0.000119	1.83696	0.005027	0.000129	
7	1.91838	0.002171	0.0000555	1.87505	0.003030	0.0000651	
8	1.93638	0.001336	0.0000278	1.90152	0.001918	0.0000347	
9	1.94910	0.0008630	0.0000149	1.92057	0.001266	0.0000194	
10	1.95839	0.0005806	0.00000836	1.93467	0.0008656	0.0000113	
11	1.96537	0.0004041	0.00000493	1.94539	0.0006101	0.00000684	
12	1.97075	0.0002895	0.00000302	1.95370	0.0004415	0.00000429	
13	1.97497	0.0002126	0.00000192	1.96028	0.0003268	0.00000277	
14	1.97836	0.0001595	0.00000126	1.96556	0.0002468	0.00000184	
15				1.96986	0.0001897	0.00000125	
16				1.97341	0.0001481	0.000000871	



## USE OF THE TABLES

In the Tables,  $C_1$  is given for unit values of  $n$ ,  $i$  and  $a$  and approaches the limit 2 as  $l$  increases. Accurate interpolation (though rarely needed—see example below) is best done with  $\log(2 - C_1)$ ,  $\log(C_3/C_1)$  and  $\log(C_5/C_1)$  as these give graphs of small curvature when plotted against  $l$  (interpolation of  $C_1$  with respect to both  $b$  and  $l$  can be achieved in this way to 1 part in 1000 on a quarto graph sheet). An example of the use of the Tables and equation (7) has already been given. As an example of equation (1), the length of a solenoid of internal radius 1 unit and external radius 3 units, which produces a field uniform to 1 part in 1000 along the axis up to 0.7 unit from the centre, will be found. From equation 1,  $[C_3/C_1](0.7)^2 = 0.001$  (the  $C_5$ -term being justifiably neglected); thus  $C_3/C_1 = 0.002$ , which corresponds to  $l = 7.2$ , so that the solenoid must be 14.4 units long.

## ACCURACY OF FORMULAE

It has been assumed in deriving the formulae that the current is uniformly distributed over the cross-section of the coil. Rosa<sup>(8)</sup> has shown that this gives good results if the wires are regularly spaced and the turns and layers are close together. Even if the winding is a rather open one, equations (1), (2) and (7) can be used with confidence in computing the initial deviations of the field from  $2\pi C_1$ . For a non-uniform winding  $C_1$  should be found as follows: the cross-section is divided into regions in which the current density is nearly uniform and  $C_1$  found for each; addition of these partial

coefficients will then give  $C_1$  for the whole coil. For example, if the solenoid consists of a number of layers of closely wound turns, the layers being separated from one another in order to allow a cooling fluid to circulate, each layer may be treated by the formula for  $C_1$  for a thin solenoid. The most accurate measurement of  $C_1$  for a thick solenoid is obtained experimentally by comparing the field with that of an accurately wound single-layer solenoid, but the computed value is a useful check.

## ACKNOWLEDGMENTS

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## Chromatic Variation of Spherical Aberration

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Formulae are given for chromatic variation of spherical aberration in an optical system, (a) considered surface by surface, (b) considered as a collection of separated thin lenses. The methods of H. Dennis Taylor, A. E. Conrady and F. D. Cruikshank are discussed and criticized.

## INTRODUCTION

All quantities, whether Gaussian or aberrational, relating to a refracting optical system, are functions of the refractive indices  $N$  of the media, and hence of the wavelength  $\lambda$  of the light traversing the system. If  $A$  is any such quantity,  $A = A(\lambda)$  expresses this functionality. In designing a system most of the calculations are done with refractive indices corresponding to a certain wavelength  $\lambda_0$  and the variations of selected quantities with wavelength are considered. Expanding  $A$  by Taylor's theorem,

$$A = A(\lambda_0) + \frac{\partial A}{\partial \lambda_0} \delta \lambda_0 + \frac{1}{2!} \frac{\partial^2 A}{\partial \lambda_0^2} \delta \lambda_0^2 + O(\delta \lambda_0^3) \quad (1)$$

The second term on the right-hand side of equation (1) is the chromatic variation of  $A$  and the third term is a

“secondary spectrum” effect; terms of third and higher degree in  $\delta \lambda_0$  are not usually important, since in practice  $\delta \lambda_0$  is small. For example, if  $A$  is the back focal length of an object-glass, the second term is the chromatic change of focus and the third the secondary spectrum in the usual sense. In this paper  $A$  is taken to be the axial spherical aberration, and the second term is the variation in this of order  $\delta \lambda$ , i.e. of order  $\delta N$ . This, the chromatic variation of spherical aberration, will be denoted by  $CVSA$ . It is necessary to emphasize that, whilst the third term is excluded from  $CVSA$ , all effects which contribute to the second term, i.e. which are of order  $\delta N$ , must be included and some of these effects are not obvious. For instance, Dennis Taylor<sup>(1)</sup> gave a formula for the  $CVSA$  of a thin lens, but did not take account of the effect of chromatic change of focus or of

the change of *CVSA* on transfer through an air-space. Conrady<sup>(2)</sup> suggested a method of obtaining *CVSA* indirectly which neglects an effect of variation in ray path, and Cruickshank<sup>(3)</sup> gave a method to which the same objection and others can be raised. The work of these last two authors is considered in more detail later.

In this paper aberration is defined in terms of optical path lengths. In Fig. 1 let *AO* be the axis of an optical system, *AP* an emergent wave-front of wavelength  $\lambda$  from an axial object point and *AP*<sub>0</sub> a sphere whose centre is at the paraxial focus of the wave-front *AP*. Let *AQ* and *AQ*<sub>0</sub> be a wave-front and paraxial sphere for wavelength  $\lambda + \delta\lambda$ . *AP* will be called the original

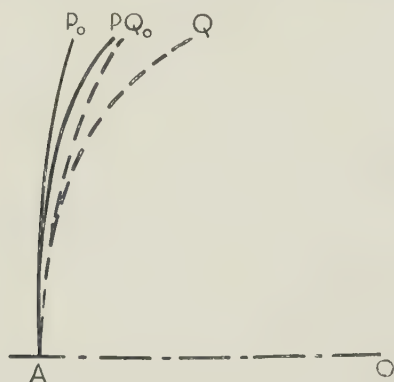


Fig. 1. Chromatic aberration of wave-fronts

wave-front and *AQ* the varied wave-front, with a corresponding terminology for rays, etc. Let square brackets denote optical path lengths, so that  $[P_0P] = N \cdot P_0P$ . If *P*<sub>0</sub>, *Q*<sub>0</sub>, *P* and *Q* lie in a line parallel to *AO*,  $[P_0P]$ , the optical path length from *P*<sub>0</sub> to *P*, is the spherical aberration of the original wave-front, and  $[Q_0Q]$  is that of the varied wave-front. For sufficiently small values of  $\delta\lambda$  (so that  $\delta\lambda^2$  is negligible),  $[P_0Q_0]$  is the wave-front aberration due to chromatic change of focus (primary chromatic aberration), and  $[PQ]$  is the total chromatic aberration, change of focus and *CVSA* included, so that  $[PQ] - [P_0Q_0]$  is the *CVSA*. These optical paths are measured parallel to the axis rather than along the normal to one of the wave-fronts or one of the spheres since it is formally necessary to choose some direction for the measurement (although the differences between all those indicated are negligible at ordinary numerical apertures); that chosen is the most convenient mathematically and it is independent of the shapes of the wave-fronts.

Explicit formulae for computing the wave-front aberration due to *CVSA* when the spherical aberration is primary are given below.

#### CHROMATIC ABERRATION AND PHASE ABERRATION

It is worth while to consider the effects of chromatic change of focus and *CVSA* on the phase relationships of the disturbances at the paraxial focus due to different

parts of the wave-front. The phase difference  $\theta$  between disturbances at the focus which originate at *A* and *P* on the original wave-front (Fig. 1) is  $(2\pi/\lambda)N \cdot P_0P$  radians, where  $\lambda$  is the wavelength in air of the disturbance and *N* is the refractive index. The corresponding phase difference  $\theta + \delta\theta$  for the varied wave-front, referred again to the paraxial focus of the original wave-front, is  $[2\pi/(\lambda + \delta\lambda)](N + \delta N)P_0Q$ . Hence  $\delta\theta$ , the change in phase difference, is

$$(2\pi/\lambda)[N \cdot P_0Q + (\delta N/N - \delta\lambda/\lambda)N \cdot P_0P]$$

for a wavelength variation  $\delta\lambda$ , neglecting quantities of order  $\delta\lambda^2$ . Of this,  $(2\pi/\lambda)N \cdot P_0Q_0$  is due to chromatic change of focus, so that the remainder of  $\delta\theta$ , which is due to *CVSA*, is

$$(2\pi/\lambda)\{[PQ] - [P_0Q_0] + (\delta N/N - \delta\lambda/\lambda)[P_0P]\}$$

Let this expression be denoted by *A* + *B*, where

$$A = (2\pi/\lambda)\{[PQ] - [P_0Q_0]\}$$

and

$$B = (2\pi/\lambda)(\delta N/N - \delta\lambda/\lambda)[P_0P]$$

It can be seen that *B* occurs because the focus is taken to be the paraxial focus of the original wave-front, and it could be made to take any other value by suitable choice of focus; for example, if the focus is taken to be the marginal focus of the original wave-front, *B* will be zero, and whatever the focus chosen *B* can be computed immediately if the spherical aberration of the system is known. The remaining term *A* is independent of the choice of focus, and is the term which is accounted for by the *CVSA* defined in the introduction in terms of optical path length; it is the term with which this paper is chiefly concerned, although in the problems of allotting optical tolerances and choosing the best state of correction (not dealt with here) account should be taken of the term *B* as well as of *A*.

#### SURFACE BY SURFACE FORMULAE FOR CHROMATIC VARIATION OF PRIMARY SPHERICAL ABERRATION

The primary spherical aberration of a wave-front is defined as in the introduction, taking account of quantities of the fourth degree in the aperture; it is taken as positive when the wave-front is in advance of the paraxial sphere, this agreeing with the usual sign convention for longitudinal spherical aberration. In symbols,

$$W = NC_4y^4 \quad (2)$$

where *W* is the primary spherical aberration, *y* is the distance from the axis to *P*, the point on the wave-front under consideration, and *C*<sub>4</sub> is the aberration coefficient for primary spherical aberration; if the wave-front is at a refracting surface, not in the space between two surfaces, *y* is equal to the paraxial incidence height at this surface of a ray through *P*, to the above degree of approximation.



Then well-known formulae give for refraction at a surface

$$\Delta(NC_4) = \frac{1}{8}\Delta\left[\frac{N}{l}\left(\frac{1}{r} - \frac{1}{l}\right)^2\right] \quad (3)$$

and for transfer from surface to surface

$$(l^4 NC_4)_{+1} = (l^4 NC_4)' \quad (4)$$

where the symbols are those usual in paraxial ray tracing (Conrady,<sup>(4)</sup> whose sign conventions are used throughout).

To find the effect of a change of refractive index it is necessary to differentiate equations (3) and (4) with respect to  $N$ . The results are

$$\Delta[\delta(NC_4)] = \frac{1}{8}Q^2\Delta\left(\frac{1}{Nl}\frac{\delta N}{N}\right) - \frac{1}{8}Q^2\Delta\left(\frac{1}{Nl}\frac{\delta l}{l}\right) + \frac{1}{4}Q\Delta\left(\frac{1}{l^2}\frac{\delta l}{l}\right) \quad (5)$$

and

$$[l^4\delta(NC_4)]_{+1} = [l^4\delta(NC_4)]' + 4\left(l^4 NC_4 \frac{\delta l}{l}\right)' - 4\left(l^4 NC_4 \frac{\delta l}{l}\right)_{+1} \quad (6)$$

where  $Q = N\left(\frac{1}{r} - \frac{1}{l}\right)$

By using equations 5 and 6,  $\delta C_4$ , the chromatic variation of the aberration coefficient  $C_4$ , could be traced through an optical system, and then  $N'\delta C_4 y^4$ , where  $N'$ ,  $\delta C_4$ ,  $y$ , refer to the last surface, would be the CVSA.

The quantities  $\delta l/l$  and  $\delta l'/l'$  which occur in equations (5) and (6) are obtained from the ordinary formula for chromatic change of focus:

$$\Delta\left(\frac{N}{l}\frac{\delta l}{l}\right) = -Q\Delta\left(\frac{\delta N}{N}\right) \quad (7)$$

For computing purposes it is preferable to transform equation (5) by multiplying by  $y^4$ ,  $y$  being the incidence height of the paraxial ray which is traced to determine  $l$ ,  $l'$ , etc. Putting  $\delta W = y^4\delta(NC_4)$ , this gives

$$\Delta(\delta W_4) = \frac{1}{8}y(Ni)^2\Delta\left(\frac{u}{N}\frac{\delta N}{N}\right) - \frac{1}{8}y(Ni)^2\Delta\left(\frac{u}{N}\frac{\delta l}{l}\right) + \frac{1}{4}yNi\Delta\left(u^2\frac{\delta l}{l}\right) \quad (8)$$

and equation (6) can be written in the form

$$(\delta W_4)_{+1} = (\delta W_4)' - 4\frac{u'd'}{y_{+1}}\frac{\delta l'}{l'}W_4' \quad (9)$$

Equations (8) and (9) are the computing equations for the quantity  $\delta W_4$ , and this quantity is actually the chromatic variation of primary spherical aberration, at the semi-aperture  $y$ .

The meaning of  $\delta W_4$  may be made clearer: referring to Fig. 1, if rectangular axes are taken with  $A$  as origin and

the  $x$ -axis along  $AO$ , the equation of the original wave-front  $AP$  may be written

$$x = \frac{1}{2l}y^2 + \left(\frac{1}{8l^3} + C_4\right)y^4 + O(y^6) \quad (10)$$

and that of the varied wave-front

$$x = \frac{1}{2}\left(\frac{l}{l} - \frac{\delta l}{l^2}\right)y^2 + \left[\frac{1}{8}\left(\frac{1}{l^3} - \frac{3\delta l}{l^4}\right) + C_4 + \delta C_4\right]y^4 + O(y^6, \delta N^2) \quad (11)$$

neglecting quantities of order  $\delta N^2$ . Then the total chromatic aberration, or optical path between the wave-fronts, is  $N\left[-\frac{1}{2}(\delta l/l^2)y^2 - \frac{3}{8}(\delta l/l^4)y^4 + \delta C_4 y^4\right]$ . The component of this which is due to chromatic change of focus is  $N\left[-\frac{1}{2}(\delta l/l^2)y^2 - \frac{3}{8}(\delta l/l^4)y^4\right]$ , and the CVSA is therefore  $N\delta C_4 y^4$ , or  $\delta W_4$  as determined by equations (8) and (9) above.

It should be emphasized that these two equations are really equations for tracing the quantity  $N\delta C_4$  through the system, and that the multiplication of equation (8) by  $y^4$  is merely a computing device which avoids the occurrence of infinite values at field lenses and enables one to use quantities such as  $y$ ,  $u$ ,  $i$ , already available from paraxial ray-tracing.

The first term on the right-hand side of equation 8 gives a change in spherical aberration due directly to the changes in refractive indices of the media, whilst the next two terms give a change due in effect to the chromatic changes in conjugates at the refracting surfaces, i.e. to the chromatic change of focus. The second term in equation (9), the "transfer" term, may be explained by reference to Fig. 2.

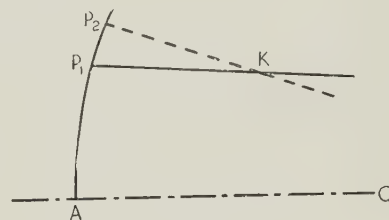


Fig. 2. Illustrating the change of CVSA on transfer

Let  $AO$  be the axis of the system and let  $AP_1$  be a refracting surface at which the wave-fronts arrive with both spherical aberration and chromatic change of focus. Let a ray of wavelength  $\lambda$  from the axial object point, which met the first surface of the system at an incidence height  $y_0$ , meet the surface  $AP_1$  at  $P_1$  and let a varied ray, i.e. a ray of wavelength  $\lambda + \delta\lambda$ , also originally incident at  $y_0$ , meet  $AP_1$  at  $P_2$ . Let these rays meet at some point  $K$ , and let the spherical aberration of the varied wave-front at  $K$  be  $W$ . Since primary spherical aberration is constant along a paraxial ray, the spherical aberration of the varied wave-front is also  $W$  at  $P_2$ ; therefore at  $P_1$ , where the original and varied wave-fronts must be compared, it is  $(y - \delta y)^4/y^4 W$ ,

where  $\delta y = P_1 P_2$ . Now since there is chromatic difference of focus,  $\delta y$  will in general\* be of the order of magnitude of  $y\delta N$  and so the increment to  $W$  will be of the order of  $\delta NW$ ; that is, it is of the order of magnitude of a chromatic variation of primary spherical aberration. Thus, where both spherical aberration and chromatic change of focus are present, the *CVSA* changes according to the position along the optical axis at which the effect is being considered, i.e. there is a change in *CVSA* on transfer as well as on refraction. It is this effect which gives rise to the last term in equation (9). It may be remarked that in the air-spaces inside a system (e.g. inside a Cooke triplet) the transfer effect is sometimes as large as, or larger than, the *CVSA* due to refraction, and it may not therefore be neglected in calculations on such systems.

#### THIN LENS EQUATIONS

It is to be expected that *CVSA*, being strictly a higher order aberration, will not be very sensitive to a change from thick to "thin" lenses such as are used in preliminary designs, and computations have shown this to be true. It is therefore worth while to develop thin lens formulae.

The choice of variables to express the conjugate distances and shape of a thin lens is a matter of some importance. Conrady<sup>(4)</sup> and Berek<sup>(5)</sup> for example, used merely one or other conjugate and radius, with the total power, and the resulting formulae are very cumbersome. Coddington gave a simpler system, quoted by Dennis Taylor<sup>(6)</sup> and Gardner,<sup>(7)</sup> in which symmetrical dimensionless functions of the conjugate distances and radii of curvature were used. However, the best system, both for primary aberrations and for the present purpose, seems to be that of Argentieri.<sup>(8)</sup> Argentieri's system is as follows: Let  $l_1$  and  $l'_2$  be the conjugate distances of a thin lens,  $r_1$  and  $r_2$  the radii of curvature of its surfaces,  $K$  its power, and  $N$  its refractive index. Argentieri's variables  $\psi$  and  $\mu$  are defined by

$$\left. \begin{aligned} \psi &= \frac{1}{l_1} + \frac{1}{l'_2} \\ \mu &= \frac{1}{r_1} + \frac{1}{r_2} - \psi \end{aligned} \right\} \quad (12)$$

One may call  $\psi$  the distance variable and  $\mu$  the shape variable. It can be seen that  $\mu$  does not specify the shape absolutely, as does the corresponding Coddington variable, but it gives the shape relative to the conjugates. More precisely, for a series of lenses of the same power but varying conjugates a constant  $\mu$  means that (for a certain incidence height  $y$ ) the algebraic sum of the angles of incidence at the two surfaces of each lens is constant.

It can easily be shown by substituting in equation (3) that the change in the spherical aberration coefficient produced by a thin lens is given by

$$\Delta(NC_4) = \frac{1}{32}K \left[ \frac{N+2}{N}\mu^2 - 2\mu\psi + \left( \frac{N}{N-1} \right)^2 K^2 \right] \quad (13)$$

or, for an incidence height  $y$ , the change in aberration is given by

$$\Delta W_4 = \frac{1}{32}y^4K \left[ \frac{N+2}{N}\mu^2 - 2\mu\psi + \left( \frac{N}{N-1} \right)^2 K^2 \right] \quad (14)$$

These formulae are considerably simpler than the others to which reference has been made. For completeness the corresponding formula for coma, in Argentieri's notation, will be given. Let  $\rho$  and  $\phi$  be polar co-ordinates in the pupil plane, let  $\eta$  be the distance from the axis of the intersection of the principal ray with the Gaussian image plane, and let  ${}_1W_3$  be the tangential coma as a wave-front aberration, so that  ${}_1W_3 = N {}_1C_3\eta\rho^3 \cos \phi$ , where  ${}_1C_3$  is the coma aberration coefficient. Then for a thin lens passed centrally by the principal ray,

$$\Delta({}_1W_3) = \frac{1}{4}N\eta y^2K \left( \frac{N+1}{N}\mu - \psi \right) \quad (15)$$

the factor  $N\eta$  being the Smith-Helmholtz-Lagrange invariant.

The remaining aberrations and stop-shift effects are found by formulae which are common to all thin lens computation systems.

It can be seen that there are only three functions of  $N$  to be tabulated for Argentieri's thin lens computing scheme (or two, if one of the two is a table of reciprocals for obtaining  $(N+2)/N$  and  $(N+1)/N$ ), whereas Coddington's scheme uses six and Conrady's eight.

To obtain the thin lens *CVSA* formulae, equation (13) is differentiated with respect to  $N$ ; the result, after some reduction, is

$$\Delta[\delta(NC_4)] = \frac{1}{32}K \left\{ \left[ \frac{N^2+2}{N^2(N-1)}\mu^2 - \frac{2}{N-1}\mu\psi + \frac{N(3N-2)}{(N-1)^3}K^2 \right] \delta N + \left[ \frac{4(N+1)}{N}\mu - 2\psi \right] \left( \frac{\delta l_1}{l_1^2} + \frac{\delta l'_2}{l'^2_2} \right) \right\} \quad (16)$$

The values of  $\delta l_1/l_1^2$  and  $\delta l'_2/l'^2_2$  are obtained from the ordinary thin lens chromatic aberration formula

$$\frac{\delta l'_2}{l'^2_2} = \frac{\delta l_1}{l_1^2} - \frac{\delta N}{N-1}K \quad (17)$$

and the transfer formula, for use when an air space occurs between lenses, is, of course, again equation (6), the values of  $C_4$  being obtained from equation (13).

As before, it is preferable to modify these equations slightly in order to facilitate computing and avoid difficulties with field lenses. Slightly different variables, defined as follows, are used:

$$\left. \begin{aligned} U &= y\mu = i_1 + i'_2 \\ V &= y\psi = u_1 + u'_2 \end{aligned} \right\} \quad (18)$$

\* Unless the whole system up to the surface considered is "thin."



using these, equation (14) becomes

$$\Delta W_4 = \frac{1}{32} y^2 K \left[ \frac{N+2}{N} U^2 - 2UV + \left( \frac{N}{N-1} \right)^2 y^2 K^2 \right] \quad (19)$$

and, on multiplying by  $y^4$ , equation (16) becomes

$$\Delta(\delta W_4) = \frac{1}{32} y^2 K \left\{ \left[ \frac{N^2+2}{N^2(N-1)} U^2 - \frac{2}{(N-1)} UV + \frac{N(3N-2)}{(N-1)^3} y^2 K^2 \right] \delta N + \left[ \frac{4(N+1)}{N} U - 2V \right] \left( u_1 \frac{\delta l_1}{l_1} + u_2' \frac{\delta l_2'}{l_2'} \right) \right\} \quad (20)$$

The transfer equation for use with equation (20) is again equation (9).

The terms in equations (20) and (9) may be divided as before into groups giving the effects of change of refractive index, change of focus, and transfer.

#### FIELD LENSES

It will be noticed that in equation (9) the transfer term can take an infinite value when transferring  $\delta W_4$  to a surface at which  $y = 0$ , i.e. where the image lies in the surface, and there is an infinite value of opposite sign on transferring from this surface to the next. This indeterminacy can be removed by means of a device suggested to the author by Dr. H. H. Hopkins for a similar purpose in connexion with higher order oblique aberrations. The device consists in referring the *CVSA* to a reference sphere at an infinite distance rather than at the surface under consideration; thus if  $d'$  is allowed to tend to infinity equation (9) becomes

$$(\delta W_4)_\infty = (\delta W_4)' + 4 \frac{\delta l'}{l'} W_4' \quad (9a)$$

giving the *CVSA* at infinity in terms of that at the particular surface to which the quantities on the right-hand side refer. If this  $CVSA_\infty$  is computed through the system it is easily seen that there are no transfer terms on going from surface to surface, but that instead there will be an extra term in equation (8), namely

$$4\Delta\left(\frac{\delta l}{l} W_4\right)$$

This can be put into a form which does not involve any indeterminacy as  $l$  tends to zero or inaccuracy for small values of  $l$ ; equation (8) will then take the following form for use with  $(\delta W_4)_\infty$ , there being no separate transfer equation:

$$\begin{aligned} \Delta(\delta W_4)_\infty = & \frac{1}{8} y (Ni)^2 \Delta \left( \frac{u}{N} \frac{\delta N}{N} \right) - \frac{1}{8} (Ni)^2 \Delta \left( \frac{u^2}{N} \delta l \right) \\ & + \frac{1}{4} Ni \Delta(u^3 \delta l) + 4W_4 \frac{N'}{r} \frac{u'}{u} \Delta \left( \frac{1}{N} \right) - 4W_4 \frac{i}{u} \Delta \left( \frac{\delta N}{N} \right) \\ & + \frac{1}{2} u' (Ni)^2 \delta l' \Delta \left( \frac{u}{N} \right) \quad (8a) \end{aligned}$$

In a similar way equation (20), for thin lenses, can be modified for use with  $(\delta W_4)_\infty$  so as to remove indeter-

minacies when the lens is at or near a focus. The modified equation is

$$\begin{aligned} \Delta(\delta W_4)_\infty = & \frac{1}{32} y K \left\{ \left[ \frac{N^2+2}{N^2(N-1)} U^2 - \frac{2}{N-1} UV + \frac{N(3N-2)}{(N-1)^3} y^2 K^2 \right] y \delta N \right. \\ & + \left[ \frac{(N+1)}{N} U - 2V \right] (u_1^2 \delta l_1 + u_2'^2 \delta l_2') \left. \right\} + 4u_2' W_4 K \delta l_2' \\ & - \frac{4W_4 Ky}{u_1(N-1)} \delta N + \frac{1}{8} y K u_2' \delta l_2' \\ & \left[ \frac{N+2}{N} U^2 - 2UV + \left( \frac{N}{N-1} \right)^2 y^2 K^2 \right] \quad (20a) \end{aligned}$$

Equations (8a) and (20a) are more complex than (8) and (20) respectively, so that although they apply to any case it is only worth while to use them in the comparatively rare cases when the image is at or near a refracting surface.

#### CONRADY'S TREATMENT OF CHROMATIC ABERRATION

Conrady<sup>(9)</sup> has discussed chromatic aberration as the difference in optical path between original and varied wave-fronts, and has given the following formula for this difference:

$$\delta W = \sum \delta N (d - D) \quad (21)$$

In this equation  $d$  is the axial path in a medium of refractive index  $N$ ,  $D$  is the path length in this medium along a trigonometrically traced ray from the axial object point and  $\delta N$  is the change in refractive index with wavelength change  $\delta \lambda$ . The summation is taken over all the dispersive media in the system, and the result  $\delta W$  is said to be the optical path between the wave-fronts at the finite aperture of the ray traced. It will be shown in this section that this formula gives the total optical path difference of order  $\delta N$  between the wave-fronts only under certain conditions.

In Fig. 3 let  $A_1 P_1$  and  $A_2 P_2$  be two successive refracting surfaces, let  $P_1 P_2$  be a ray from the axial object point

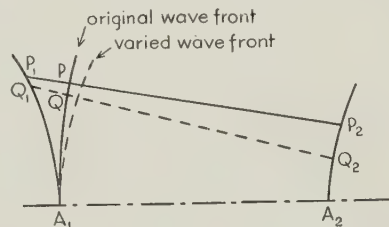


Fig. 3. Conrady's treatment of chromatic aberration

(a true ray, not paraxial) and let  $Q_1 Q_2$  be the corresponding varied ray, which coincided with the original ray in the object space. Let  $P_1 P_2 = D$ ,  $Q_1 Q_2 = D + \delta D$ ,  $A_1 A_2 = d$ . Then Conrady's argument is as follows:

The optical path difference between the marginal and axial rays from object to image is  $W = \sum N(d - D)$ .

Hence for a variation  $\delta\lambda$  in wavelength the change in this optical path difference is

$$\delta W = \Sigma[\delta N(d - D) + N\delta(d - D)]$$

But by Fermat's theorem  $\Sigma N\delta(d - D)$  is of order  $\delta N^2$  when the values of  $D$  refer to an actual ray path, so that

$$\delta W = \Sigma\delta N(d - D) \quad (21, \text{bis})$$

Now in this argument it must be noticed that what is found is the change in optical path difference from the original ray path  $\dots P_1P_2 \dots$  to the varied path  $\dots Q_1Q_2 \dots$ . But by a similar argument to that used at the end of the section on surface by surface formulae it may be shown that this quantity differs from the optical path difference between the original and varied wave-fronts at a particular semi-aperture by an amount which is in general of the order of magnitude of  $CVSA$ . For, if  $W$  is the spherical aberration of the varied wave-front at  $Q$ , its value at  $P$ , where the two wave-fronts are to be compared, will be  $(y + QP)^4/y^4 W$ , if the spherical aberration is purely primary, and since  $QP$  is generally of order  $y\delta N$  the change is of order  $W\delta N$ , i.e. of the order of magnitude of  $CVSA$  unless  $W$  happens to be zero.

This shows that in a space where chromatic change of focus causes a first-order chromatic variation in incidence height of the ray, i.e. in general in an optical system, Conrady's formula only gives the optical path between the original and varied wave-fronts correct to quantities of order  $y^2\delta N$ , or, of the order of chromatic change of focus; hence the difference between the chromatic change of focus and  $\delta W$  from equation (21) does not give the  $CVSA$  as suggested by Conrady.<sup>(2)</sup> However, it can easily be seen that if the above condition occurs in the middle of an optical system but the varied and original rays unite again on emerging from the last surface, Conrady's formula will give the total optical path between the two wave-fronts correct to, i.e. including,  $CVSA$  after the last refraction, although not in intermediate spaces. The condition that the rays shall re-unite at the last surface is, of course, substantially the condition that the system shall be corrected for chromatic difference of magnification, and this is the case for the majority of systems after an early design stage (but not for all systems; spectrographs, for example, are often quite uncorrected chromatically, but it is desirable to eliminate  $CVSA$ ); hence for such systems Conrady's formula will give the  $CVSA$  at finite aperture in the image space but not in intermediate spaces, and the condition  $\Sigma\delta N(d - D) = 0$  will ensure good zonal chromatic correction.

It is not possible to obtain a "primary"  $CVSA$  formula for use with a paraxial instead of a finite ray-trace from Conrady's condition, even with the above restrictions on validity. To see this, let  $D$  be the distance along the finite ray from surface to surface and  $\underline{D}$  this distance for the corresponding paraxial ray. Then the angular spherical aberration of the finite ray is of order  $y^3$ , and

hence  $D - \underline{D}$  is of order  $y^4$ . Thus the difference between  $\delta N(d - D)$  and  $\delta N(\underline{d} - \underline{D})$  will affect  $CVSA$ . Therefore equation (21) can only be "paraxialized" to give the chromatic variation of focus.

#### CRUICKSHANK'S METHOD FOR $CVSA$

Cruickshank<sup>(3)</sup> describes a method of obtaining  $CVSA$  at finite aperture from a paraxial and a marginal (or zonal) ray trace; this is based on his system of transfer coefficients described in a previous series of papers.<sup>(10, 11)</sup> According to this method  $\partial LA'/\partial N$ , the partial derivative of the longitudinal spherical aberration with respect to refractive index, is obtained for each component of a system by recurrence formulae involving the data of the ray-traces.

There are two serious criticisms to be made of this method:

(i) The quantity obtained gives no information at all about  $\partial W/\partial N$ , the derivative of the wave-front aberration, since  $\partial LA'/\partial N$  is the derivative of  $\partial W/\partial N$  with respect to the aperture (apart from a constant factor). It is, in fact, easy to give examples such as might well occur in practice where  $\partial LA'/\partial N$  is zero and  $\partial W/\partial N$  is not, or vice versa. Fig. 4 shows the dispositions of the wave-fronts and rays in such cases.

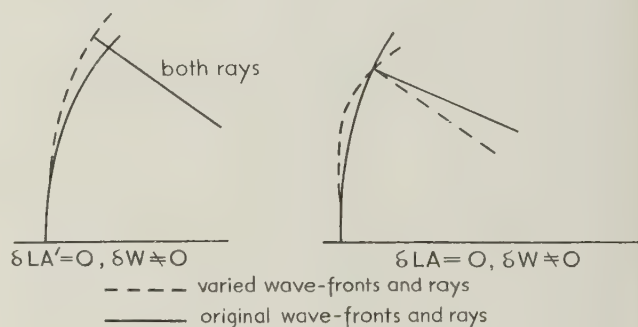


Fig. 4. Comparison of ray and wave-front methods of treating  $CVSA$

(ii) The method is valid only in the same cases as Conrady's method, i.e. when the varied ray emerges from the same point as the original ray or when there is no spherical aberration in the wave-front. For, if  $\delta LA'$  is the chromatic variation of longitudinal spherical aberration,  $\delta LA'$  is of the order of magnitude of  $y^2\delta N$ . But the chromatic change in incidence height is of order  $y\delta N$  and this causes a further shift of the ray intersection of order  $y^2\delta N$ , i.e. of the same order of magnitude as  $\delta LA'$ .

It is impossible to derive an expression for  $CVSA$  at finite aperture applicable to a single marginal (or zonal) ray trace which shall be valid whatever the state of correction of the system. The reason for this lies in the nature of the correcting term for the effect of the varied ray path. As explained previously in the Section



on Conrady's treatment this term is of the form  $(y + PQ)^4/y^4W$ , where  $PQ$  is the change in incidence height of the varied ray, when  $W$  is primary spherical aberration; however, for secondary spherical aberration it would be of the form  $(y + PQ)^6/y^6W$ , and so on for higher orders. But since in a system of moderate aperture there are appreciable amounts of secondary and tertiary spherical aberration, and a single marginal (or zonal) ray trace gives no information about the relative proportions of these, it is impossible to give any definite formula for this correcting term.

#### A NUMERICAL EXAMPLE

An example will now be given of some values computed by means of equations (8) and (9) for an actual lens specification; this will show the relative sizes of the various contributions to  $\delta W_4$  which can occur. The specification was supplied by Kodak Ltd., to whom thanks are due. It was of a 20 in F: 6.3 objective of the Taylor triplet type. The primary CVSA was computed at full nominal aperture for the spectral range  $C$  to  $F$ , and the results for each surface are given in the table. The quantities  $A$ ,  $B$  and  $C$  are respectively the first term on the right-hand side of equation (8), the sum of the second and third terms, and the second term on the right-hand side of equation (9). Thus  $A$  is the contribution to  $\delta W_4$  from refractive index change,  $B$  is that from change of focus and  $C$  is the transfer term. All are expressed in microns of optical path.

It is interesting to note that the  $A$ 's have the least effect at most surfaces, and that the  $B$ 's, giving the effect on spherical aberration of change of conjugates, are usually most important. The air spaces, after surfaces 2 and 4, are about 1.6 and 2.2 in respectively, and the  $C$ 's for these spaces are quite high.

Table of values computed for an actual lens specification

Surface	1	2	3	4	5	6
$A$	0.095	-0.159	0.155	-0.046	0.000	-0.202
$B$	-0.035	3.190	-6.780	-1.038	-0.005	0.193
$C$	0.021	1.500	0.009	1.611	0.244	0
Final $\delta W'_4 = -0.843$						
Final $W'_4 = 4.2$						

#### ACKNOWLEDGMENTS

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## Vacuum Factor of the Oxide-Cathode Valve

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The vacuum factor  $k$  of an oxide-cathode triode valve is defined as the positive ion current measured in a negative control-grid per milliamperere of ionizing electron current to the anode. Measurements on a variety of new valves show wide variations of  $k$ , but these values fall, after a period of operation, to an approximately constant value  $k_0$  in the range 300 to 900  $\mu\text{mA}/\text{mA}$ . The characteristics of this residual vacuum factor  $k_0$  are examined in some detail and a theory proposed to explain the anomalous form of variation of  $k_0$  with anode voltage. This theory has a bearing on the interpretation of ionization gauge measurements at pressures below  $1 \times 10^{-6}$  mm. of mercury.

It is commonly supposed that the life of the high-vacuum oxide-cathode valve is a function of the quantity of residual gas left in the tube after production processing. Measurement of the residual gas pressure is therefore a matter of considerable interest and is always attempted by arranging the valve to act as its own

ionization gauge. Since the pressures in such valves are normally below  $10^{-4}$  mm. of mercury the ionization method is a convenient one in that the number of positive ions formed from the residual gas is directly proportional both to the pressure of the residual gas and to the density of ionizing electron flow. Measurement

of the number of positive ions formed is achieved by attracting them to a negatively-primed electrode and determining the electron flow required to keep this electrode neutral. The magnitude of the positive ion flow per mA of ionizing electron current is thus a measure of the residual gas pressure and is known as the vacuum factor of the valve.

There appears to be little systematic work on the subject of the vacuum factor apart from a paper by Herrmann and Krieg<sup>(1)</sup> in which the factors of a number of commercial valves were measured and compared. The results showed wide variations of magnitude between valves of different types. The object of the present paper is to describe more recent work on the general characteristics of the vacuum factor and to attempt to systematize the subject in a way that has not yet been achieved.

#### TECHNIQUE OF MEASUREMENT

*General Principle.*—Determination of the vacuum factor of a triode valve consists of a measurement of the positive ion flow to the negatively-primed control grid when 1 mA of electron current is passing to the anode whose potential is of the order of 150 to 300 V. For residual gas pressures below  $10^{-3}$  mm. of mercury the number of ionizing encounters is directly proportional both to gas concentration and density of the electron stream. Thus if

$I_{RG}$  = Positive ion flow (usually known as reverse grid current);  $I_A$  = Anode electron current; and  $p$  = Gas pressure

$$\begin{aligned} \text{then, } I_{RG} &\propto p; \\ &\propto I_A; \\ &= CpI_A \end{aligned}$$

$$\text{or, } I_{RG}/I_A = Cp = k \quad (1)$$

where  $k$  is the vacuum factor appropriate to the pressure  $p$ .  $C$  is a proportionality constant depending on the geometry of the valve structure and to the nature of the residual gas. This constant can be determined experimentally for a particular geometry by observing the vacuum factor  $k$  in a valve connected to a McLeod gauge working in the range  $10^{-3}$  to  $10^{-4}$  mm. of mercury. Since the bulk of the gas in a high-vacuum valve is normally in the adsorbed state on internal surfaces and only moves under thermal or electrical disturbance, the conception of a pressure in the accepted sense is probably unsound. In the present paper therefore the quantity of gas will be presented in the form of the vacuum factor measured in micro-microamperes of positive ion current per milliampere of electron current.

*Precautions to be taken in Measurement.*—Two major sources of error were encountered in the measurement of  $I_{RG}$ . The first and most serious was insulation leakage between anode and control or ion collector grid. This was overcome by mounting the anode on a glass insulating pillar and taking the anode connexion out

through the glass envelope of the valve at the end remote from that at which the control grid was led out. This precaution was augmented by the use of "guard rings" at the base of the anode supporting pillar and on the internal and external surfaces of the glass envelope. With these arrangements leakage from anode to control grid was less than  $10 \mu\mu\text{A}$  with anode at 400 V potential. The other source of error encountered was electron emission from the control grid. This emission must be eliminated as far as possible since it is indistinguishable in the external grid circuit from a positive ion flow. The emission may be thermal or photoelectric. The thermal component can be readily measured in the grid-cathode circuit and eliminated by reducing the grid temperature. The photoelectric component is due either to visual light or to soft X-rays originating at the electron bombarded anode. The component due to visual light can be eliminated by excluding daylight from the envelope and slightly underrunning the oxide cathode. Little can be done at this stage about the X-ray photo-emission and more will be said of its effects later in the paper.

Measurement of the positive ion current presents some difficulty as it is normally in the range  $10^2 \mu\mu\text{A}$  to  $10^5 \mu\mu\text{A}$ . A two-metre reflecting galvanometer with a sensitivity of  $20 \mu\mu\text{A}/\text{mm.}$  was used in the present work and found adequate. With the employment of high anode voltages (50 to 400 V) it will be apparent that the measurement of such small currents required a test set of high insulation properties. With the aid of "guard rings," however, these insulation difficulties were overcome. The test circuit employed was a conventional triode one with meters for measurement of anode current and voltage, negative grid voltage and heater voltage.

#### CHARACTERISTICS OF VACUUM FACTOR $k$

*Preliminary Measurements.*—Preliminary measurements were made on a series of commercial pentode receiving valves of different types and selected in the laboratory for absence of anode-grid mica insulation leakage. The valves were arranged as triodes with anode, suppressor and screen grids connected as a common "anode" maintained at 200 V. The values of  $k$  obtained varied from  $2000 \mu\mu\text{A}/\text{mA}$  to  $500,000 \mu\mu\text{A}/\text{mA}$ . Two factors of significance were, however, noted during the measurement. Firstly, the valves of a similar type from a common source had values of  $k$  which were fairly closely grouped. Secondly, all valves showed  $k$  to be unstable with time; some increasing but the majority decreasing in magnitude.

*The Vacuum-Factor/Time Characteristic.*—The characteristic of  $k$  against time  $t$  was next investigated. A valve was set as a triode with 200 V on the anode and a suitable negative voltage on the control grid. These potentials were maintained constant and the positive ion current in the control grid measured over an appropriate period. In general, all valves tested showed the same form of  $k/t$  characteristic—a rapid and smooth fall of  $k$



in the early stages gradually slowing up to make an asymptotic approach to a steady low value. Differences between valves showed up as a difference in time to approach the final value of  $k$ . As might be expected, valves with a high value of  $k$  at time  $t = 0$  showed a longer time to achieve the final value of  $k$ . The main variant from this generality was a tendency to irregularity in the rate of change of  $k$  in the earliest stage of the test. Some valves showed an increase in  $k$  which rose to a maximum and then fell in the normal manner. This is probably an effect of secondary importance and due to outgassing of the anode during the warming-up period. A typical result is shown in Fig. 1. The first and most interesting observation was that all valves fell to

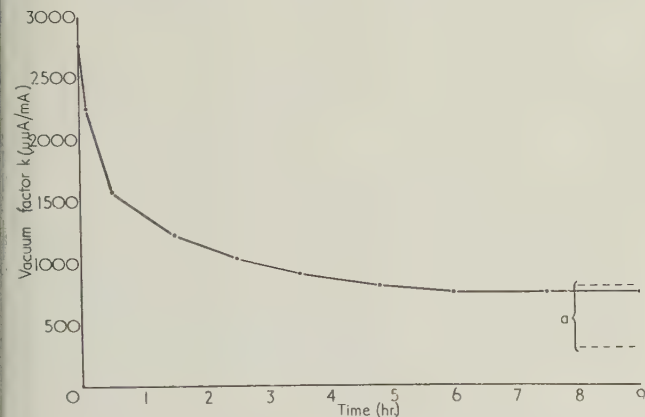


Fig. 1. Typical example of vacuum factor/time characteristic (a)  $k_0$  range at  $E_A = 200$  V for all valves tested

a final value of  $k$  within a relatively narrow margin (300 to 800  $\mu\text{A}/\text{mA}$ ) irrespective of the initial values which spread over the range of 2000 to 500,000  $\mu\text{A}/\text{mA}$ . This result was surprising as the valves tested were pentodes and triodes of different electrode geometries and from different manufacturing sources. Special triodes made in the laboratory gave similar results. The only common feature of these tests was an anode potential of 200 V. The second point of interest was the stability of  $k$  once it had reached its steady low value. Measurement over a period of 250 hr. produced no changes greater than those explainable by the experimental difficulty of measuring the small positive ion current. The third interesting feature of the  $k/t$

characteristic is that the integral  $\int_{t_0}^t f(k) dt$  gives a direct means of comparing the number of ionic collisions that occur in the valve before  $k$  reaches its steady low value. The fall of  $k$  is due, of course, to absorption of the positive ions either on the negative grid surface or in the oxide-cathode which is maintained at earth potential and is also attractive to positive particles. Some ions reaching the negative grid will be neutralized and escape to become available for a further cycle. As  $k$  approaches its final value the bulk of the

gas will probably have found its way into the massive oxide cathode from which further escape is difficult. The integral thus gives some idea of the degree of gas poisoning to which the cathode is subjected.<sup>(2)</sup> As a first approximation it will, therefore, be concluded that the vacuum factor (or pressure) of the high-vacuum oxide-cathode valve falls to a low constant value when the valve is run under ionizing conditions. This value is in the range 300 to 800  $\mu\text{A}/\text{mA}$  and may indicate a common final operating pressure for such valves. The value will be described as the residual vacuum factor  $k_0$ .

#### CHARACTERISTICS OF THE RESIDUAL VACUUM FACTOR $k_0$

*Variation of Efficiency or Probability of Ionization with Electron Energy.* The number of ions produced per milliamper of electron flow increases from zero at the ionizing potential to a maximum in the electron energy range 90 to 120 eV and thereafter declines slowly and apparently indefinitely. All the common gases follow this general characteristic. A typical example from Compton and Van Voorhis<sup>(3)</sup> for oxygen is shown as Curve A in Fig. 2. It was pointed out by Herrmann and

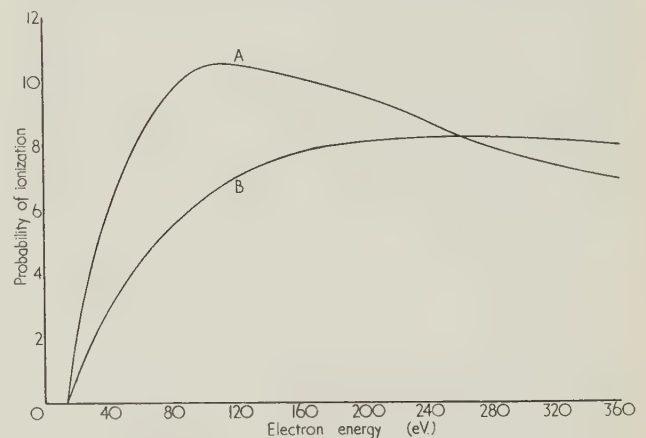


Fig. 2. Characteristic of ionization probability/electron energy. Curve A, constant velocity electron stream; Curve B, constant acceleration electron stream

Runge<sup>(4)</sup> that the normal probability characteristic is only applicable to a constant-velocity electron beam and that the accelerating stream in a triode valve requires further consideration before a probability characteristic can be derived. Assuming a constant acceleration over the whole electron path a direct integration of Curve A will lead to the required characteristic. Such an integration has been undertaken graphically and leads to a characteristic of the form Curve B. The maximum has shifted to an anode voltage of about 250 V and the characteristic is very flat between 200 to 300 V. Tests on a tungsten-filament triode confirmed the prediction at a pressure of about  $10^{-3}$  mm. of mercury.

*The  $k_0/E_A$  Characteristic.* The variation of residual vacuum factor  $k_0$  with anode voltage  $E_A$  was next

investigated for a series of the specially prepared oxide-cathode triodes previously described. The control-grid voltage was maintained at a constant negative voltage and the anode voltage varied. Values of  $k_0$  were obtained over the anode voltage range 70 to 200 V. During this series the anode wattage increased with each increase of anode voltage. Two results shown in Fig. 3 are typical of a total of more than twenty valves tested. In general there is an approximately linear relationship in the range 100 to 200 V and a flattening of the characteristic below 100 V. All valves show this transition within the range 100 to 130 V. Individual valves were tested up to 400 V and showed that linearity was maintained at least up to this limit. At this stage the measurements were repeated with a variant. Each increase in anode voltage was accompanied by a sufficient increase in negative grid voltage to maintain a constant anode wattage. No change in the  $k_0/E_A$  characteristic was observed. It is apparent that there is a wide difference between the  $k_0/E_A$  characteristic and its predicted form set out as Curve B in Fig. 2. An attempt to reconcile the difference will be made in the next section.

*The  $k_0/E_H$  Characteristic at Constant  $E_A$ .*—Variation of  $k_0$  with the temperature of the oxide-cathode was the next point to be investigated. The cathodes were the common indirectly-heated type with mixed barium-oxide/strontium-oxide on a nickel core. The heater voltage was 6.3 V (or a nominal cathode temperature of 1000° K.). The value of  $k_0$  was examined at  $E_A = 140$  V over a range of heater voltage from 3.5 to 6.5 V, with the following results:

Heater voltage	Approximate cathode temperature	$k_0 \mu A/mA$
3.50 V	870° K.	152.0
4.00 V	930° K.	151.6
4.50 V	970° K.	152.1
5.00 V	1010° K.	151.6
5.50 V	1050° K.	150.9
6.00 V	1080° K.	151.4
6.50 V	1120° K.	152.6

The residual factor  $k_0$  is therefore invariant with the temperature of the oxide-cathode. For heater voltage higher than 7 V there is a rapid increase in reverse grid current but this was found to be due to primary electron emission from the overheated grid.

*Influence of Envelope Temperature on  $k_0$ .*—Increasing the envelope temperature of a valve by 25° C. showed an increase of vacuum factor but this gradually returned to the residual value  $k_0$ . Presumably this rise is due to adsorbed gas leaving the warm glass surface. Absorption of this gas in the cathode results in the vacuum factor returning to  $k_0$ . Cooling the envelope produced no measurable change in  $k_0$ . The cooling was effected by lowering a special valve into freezing alcohol and then into liquid oxygen. The tube was maintained at the lower temperature for 30 min. but no change of  $k_0$  was noted. During these measurements the heater

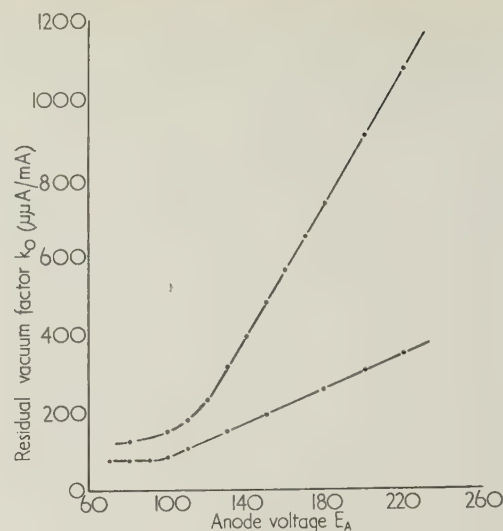


Fig. 3. Typical  $k_0/E_A$  characteristics of oxide-cathode valves.

voltage was maintained at 6.3 V and it was observed that the anode current fell slightly. This was doubtless due to the radiation cooling of the cathode by the surrounding envelope at  $-183^\circ\text{C}$ . This invariance with envelope temperature is informative and shows that  $k_0$  cannot be ascribed to barium metal vapour or to a whole range of gases with boiling points higher than that of liquid oxygen.

*Summary.* To summarize, it appears that  $k_0$  is:

- (a) a direct function of anode voltage  $E_A$
- (b) independent of anode current
- (c) independent of anode temperature providing no out-gassing occurs
- (d) independent of oxide-cathode temperature
- (e) independent of envelope temperature providing adsorbed gas is not evolved.

The only variable on which  $k_0$  appears to be dependent is therefore the energy possessed by the electron when it strikes the anode.

*The  $k_0/E_A$  Characteristic of a Tungsten Filament Triode.*—Since the value of  $k_0$  for the oxide-cathode valve appears unrelated to the nature of the electron emitter it follows that a similar form of  $k_0/E_A$  characteristic should be obtained from a tungsten filament triode. Furthermore the transition range (100 to 130 V) should remain unchanged. Experiments were undertaken with such a tube which was fitted with a cylindrical nickel anode, a molybdenum helical grid and a straight tungsten-filament cathode. After thorough processing, the valve was sealed at pressure of  $2 \times 10^{-6}$  mm. of mercury. Subsequent tests showed a close similarity in behaviour between the tungsten and oxide-cathode cases. The vacuum factor  $k$  fell with time to a final steady residual factor  $k_0$ . The form of the  $k_0/E_A$  characteristic was very similar to that of the oxide-cathode case and four experimental tubes all showed a transition effect in the



range 100 to 130 V. A typical result is shown as Curve A in Fig. 4.

A search of the literature at this stage showed that almost identical results were obtained by Jaycox and Weinhart<sup>(5)</sup> in 1931 for a thoriated tungsten filament triode. This result is plotted as Curve B in Fig. 4 and

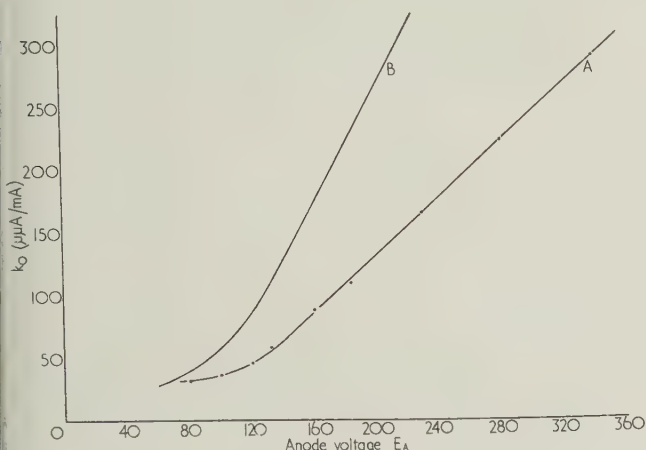


Fig. 4.  $k_0/E_A$  characteristic of tungsten-filament triodes

shows a transition effect at 120 V. The reduction of  $k$  to  $k_0$  in the case of the tungsten filament is probably due to residual gas absorption on the negative grid and to chemical reaction with the tungsten filament.

#### DISCUSSION OF RESULTS

The striking difference in form of the  $k_0/E_A$  characteristic and the "probability"/ $E_A$  characteristic of Fig. 2 remains for discussion. The probability curve is another form of the  $k_0/E_A$  characteristic and should differ from it only in respect of the units employed for the ordinate axis. The difference between the two forms is explained in the following manner. The  $k_0/E_A$  characteristic obtained in the present work is regarded as the resultant of two superimposed characteristics—one being of the normal probability form and the other a linear characteristic with an intercept at 100 to 130 V. This conception is shown in Fig. 5. The component with normal probability form is regarded as due to a true positive ion flow resulting from residual gas collision processes. The linear component is probably due to a loss of electrons by the negative collector under irradiation by soft X-rays. Such X-rays are generated at the surface of the bombarded anode and their ability to release electrons from the negative collector may be regarded as a function of  $E_A$ .

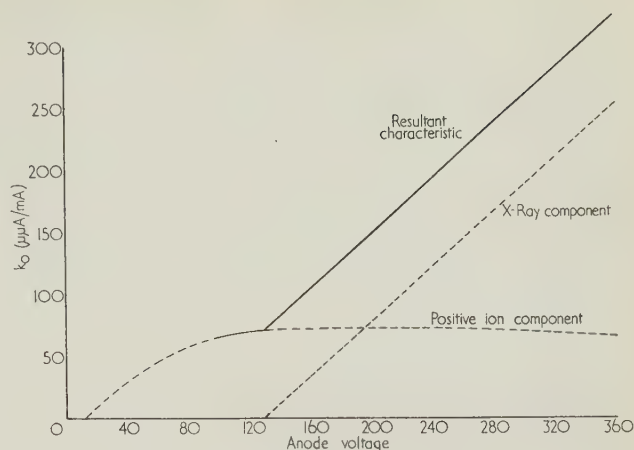


Fig. 5. Compounding of characteristics

A possible alternative to the X-ray theory exists and cannot be ruled out at this stage. The linear characteristic might be due to a true positively ionized stream of metal ions torn from the electron bombarded anode surface. It is conceivable that the number of such sputtered atoms or ions might be a function of the energy of electron impact. A final point may be made concerning the influence of electrode geometry on the general form of the  $k_0/E_A$  characteristic. Experiments with a number of geometries in the oxide-cathode case showed no change. Some recent work by the author's colleague, Dr. S. Wagener, employed a tetrode valve with a probe wire collector fitted close to the anode and his findings were a normal  $k_0/E_A$  characteristic with a transition at 100 V. The general form of the characteristic is therefore independent of valve geometry.

#### ACKNOWLEDGMENTS

The author wishes to express his appreciation to the Engineer-in-Chief of the Post Office for permission to publish this work and also to Mr. H. Batey for his skilled technical assistance.

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## NOTES AND NEWS

## Correspondence

## The Exchange of Diagrams and Data between Radiation Therapy Centres

A scheme organized by the Hospital Physicists' Association to facilitate the exchange of useful diagrams and data between radiotherapy centres has already been described.\* Briefly, it was felt that most centres accumulate isodose and other charts, nomograms, machine drawings, and tabulated data which would often be useful to many other centres. Furthermore, diagrams in the literature are generally too small to be of practical use, and large copies of the originals might also be useful (proper precautions being taken to comply with copyright law). Therefore, catalogues have been issued by the Hospital Physicists' Association, describing all the material available of which copies can be supplied by the honorary Secretary of the scheme. Catalogue A, issued in December 1944, contained 213 items, and catalogue B, of June 1946, over 200 more. By this time some 350 diagrams, etc., had been supplied, some of which went to the U.S.A., Canada, and New Zealand. By the end of 1949 the enquiries about the scheme and requests for catalogues, particularly from abroad, showed no signs of abatement, and about 400 copies of the first two catalogues had been distributed as follows:—

British Isles	52 per cent.
U.S.A.	26 "
Canada	7.5 "
Europe (excluding British Isles)	6 "
Australasia	3 "
India	2.5 "
South Africa	2 "
South America	1 "

The total number of diagrams, etc., now distributed exceeds 2 500, 45 per cent. of which have gone abroad. Within a single recent period of 12 months, nearly 900 items were despatched.

Enough new material has accumulated since the issue of catalogue B to justify the production of catalogue C (December 1949). This contains 270 items classified by the number system used previously. Its scope is illustrated by the Table:—

1. *X-rays*

- 1.1. Isodose curves, and diagrams, etc., relating directly to X-ray therapy, e.g., backscatter factors, depth doses:—
  - 1.1.1. Isodose curves of single fields 157
  - 1.1.2. Isodose curves illustrating the summation of fields 43

\* READ, J. *Brit. Journ. Rad.*, **18**, p. 116 (1945); *Brit. Med. Bull.*, **4**, p. 64 (1946).

- 1.1.3. Diagrams and data relating directly to X-ray therapy, e.g., backscatter factors, depth doses 5
- 1.2. Wedge field and other non-standard isodose curves 14
- 1.4. Physical data relating solely to X radiation:—
  - 1.4.2. Data relating to half-value layers 1
  - 1.4.3. Miscellaneous 2
- 2. *Radium and Radon*
  - 2.2. Isodose curves, and rules, graphs, etc., for dose calculation for radioactive sources:—
    - 2.2.1. Linear sources,  $\gamma$  rays 11
    - 2.2.5. Miscellaneous sources,  $\gamma$  rays 16
  - 2.4. Data relating to radon 1
- 3. *Physical Data*
  - 3.1. Scattering and absorption data for radiation in general:—
    - 3.1.1. Data relating to scattered radiation based on Klein-Nishina and Compton formulae 2
    - 3.1.3. Miscellaneous absorption and transmission data 10
  - 3.2. Energy absorption, properties of the ionizing particles, distribution of ion pairs 2
- 4. *Mathematical Data* 2
- 5. *Machine Drawings*
  - 5.1. Ionization chambers 3
- 7. *Reprints of which Stocks are exhausted* 1

The scheme has been run so that it is just self-supporting financially. The accumulation of negative copies of diagrams has made possible reductions in charges so that the average cost is now about 3s. 2d. (£0.16) per item, compared with 4s. 5d. (£0.22) in 1945. The majority of X-ray isodose charts may now be obtained on paper for about 2s. 0d. (£0.10) each.

Facilities have been granted to the Hospital Physicists' Association to demonstrate the working of the scheme at a stand in the Scientific Exhibition Section of the forthcoming 6th International Congress of Radiology in London next July. It is hoped that visitors, particularly those from overseas, will profit from this opportunity to obtain first-hand information regarding the service provided.

It is desired to thank all those whose willing co-operation has made possible this effort to serve the interests of radiation therapy. The Association owes a special debt of gratitude to the British Empire Cancer Campaign, whose timely gift of £50 alleviated the initial financial difficulties.

Copies of catalogue C can be obtained (5s. or £0.25, post free) from the undersigned.

D. E. A. JONES,  
Honorary Secretary,

Mount Vernon Hospital,  
Northwood, Middlesex.

Diagrams and Data Sub-Committee  
Hospital Physicists' Association

## New Books

**English for Science Students.** By P. A. GASKILL, M.A., and A. TOWERS, M.Sc. (London: J. M. Dent and Sons.) Pp. xii + 178. Price 2s. 9d. net.

This is an attempt by two schoolmasters, one a specialist in English and the other in Science, to provide a shortened

course in English Composition for science students. It consists, in the main, of a number of well-chosen extracts from the writings of various authors, which the reader is invited to study and comment upon. An introductory section deals with points of grammar, spelling and composition on which inexperienced writers are most apt to go astray and



a glossary of grammatical terms completes the work. The book provides an excellent basis for a course in English for, say, a Science Sixth Form, and its general adoption in the schools might do much to correct the deplorable inability shown by so many students to write their own language correctly or even intelligibly. The adult worker, who can no longer command the services of a kindly mentor, might find the book somewhat difficult.

J. A. CROWTHER

**Deformation and Flow.** An Elementary Introduction to Theoretical Rheology. By M. REINER, D.Tech. (London: H. K. Lewis and Co., Ltd.) Pp. xviii + 346. Price 32s. 6d. net.

It is rather difficult to indicate briefly the scope of this book—its themes are so many and varied. It is in no sense a general survey of rheology, but is intended rather as an elementary introduction to the more theoretical aspects of the subject. There is less formal mathematics than in the author's earlier *Ten Lectures in Theoretical Rheology* and correspondingly more emphasis is placed on physical interpretation. Subjects dealt with include: the flow of Newtonian and non-Newtonian liquids and suspensions in capillary and concentric cylinder type instruments; the mathematical formulation of rheological properties (with a discussion of power-law relations); the implications of Trouton's experiments on flow under tensile stress, with particular reference to the significance of "volume viscosity"; the work-hardening and tensile properties of steel, illustrated by some illuminating experiments by the author; criteria for plastic yield and rupture; problems in bending and torsion; elastic after-effects and creep in concrete; and the analysis of complex types of behaviour.

In the preparation of this book more thought might well have been given to the arrangement of the material, with a view to achieving a more systematic and smoothly flowing story. As it stands, one finds perfectly straightforward general physics interspersed with highly debatable (and at times metaphysical) speculation in a rather disturbing manner. A number of interesting experiments are reported, but the conclusions drawn do not always carry conviction and some of the mathematical arguments require rather more than an elementary understanding on the part of the reader. For example, commenting on the observation that the flow of concrete mixes obeys the Einstein equation for dilute suspensions of spherical particles for filler contents as high as 60%, the author proceeds, by an argument of very doubtful consistency, to conclude that this concentration of solids in a medium of very high viscosity is equivalent to an infinitesimally small concentration in a low-viscosity liquid. One wonders whether, on this basis, a 99.9% suspension should likewise conform to the Einstein equation! Again, in dealing with relatively concentrated rubber solutions in terms of the Einstein equation, no reference is made to the statistical concept of long-chain molecules developed during the last ten years and its bearing on the structure and properties of polymer solutions. In these circumstances the discussion of "structural" viscosity seems a little unreal.

Finally there is the vexed question of nomenclature. We are asked to believe that paint, which can be poured from a can, is a solid whilst concrete is a liquid. Glass cannot yet be assigned a definite status because of the uncertainty in the estimates of its viscosity—these ranging from  $10^{19}$  to  $10^{60}$  poises. Rheology may thrive on paradoxes, but is it

really necessary to get so far away from the ordinary meanings of words? It is to be hoped, however, that physicists will read this book, for it is pervaded throughout by the author's lively imagination and deep interest in fundamental problems. It can hardly fail to arouse curiosity and stimulate further investigation in a field where much spade work remains to be done.

L. R. G. TRELOAR

**Atomic Energy Year Book.** Edited by DR. J. TUTIN. (London: Temple Press Ltd.) Pp. xx + 237. Price 21s. net.

This book is intended "... to provide a convenient means whereby the industrialist and technician may have at hand reasonably up-to-date information on the development of atomic energy." The chapters concerned with the history of the subject and with the sale, distribution and handling of radioactive isotopes are good, although there is no reference to British work on atomic energy prior to "Tube Alloys." But the rest of the volume, dealing with actual and potential developments in the atomic energy field and with the results of medical research, is, in the reviewer's opinion, not well balanced and sometimes inaccurate. For example, although the Canadians possess, in the N.R.X. pile, the most powerful known reactor outside the U.S.A., it is specifically stated on p. 85 that their only pile is the "zero energy" pile known as Zeep. Any succeeding issue of this Year Book will need to be better co-ordinated and more critically compiled if it is fully to accomplish its stated purpose.

J. THEWLIS

**Transactions of the Spectacle Makers' Company Convention, 1948.** (London: Worshipful Company of Spectacle Makers.) Pp. iv + 111. Price 21s. net.

This volume contains technical papers on ophthalmic optics presented by such well-known authors as W. Swaine, A. G. Bennett, Lord Charnwood, W. D. Wright, H. C. Binstead, C. S. Flick, H. Hartridge and W. S. Stiles at an Optical Convention held in November 1948 to mark the Fiftieth Anniversary of the establishment of Examinations in Visual Optics by the Worshipful Company of Spectacle Makers. In an introductory paper, Sir William Champness reminds us of the important part played by the Company in the field of education since its establishment in 1629. The paper by A. J. Philpot on "The Need for Research and the Means of Meeting It" gives an appropriate indication of the next stage of advance. The whole production commemorates in a worthy manner a significant development in technical training.

H. LOWERY

**Metals and Alloys.** (London: The Louis Cassier Co., Ltd. Distributed by Iliffe and Sons Ltd.) Pp. 214. Price 15s. net.

This, the fifth revised edition, contains the composition of some 4 600 non-ferrous alloys as compared with 3 700 in the fourth edition, many of which have been rejected in the new version. The alloys covered are limited to those containing not more than 50% of iron. Alloys included are those with specific "names" and in addition many others inserted in their classes (brass, bronze, etc.). The list contains much practical information and is a useful addition to the library or laboratory bookshelf. An omission is the Alcoa (Aluminum Co. of America) series of alloys: reference to

these in the literature is nowadays very frequent. An addition which suggests itself is names of manufacturers of the alloys mentioned, but while this is feasible for "named" alloys, in other cases it would perhaps lead either to long lists of names or to invidious distinctions: no doubt it is some such consideration as this which has led to the exclusion of the manufacturers' names.

G. L. J. B.

**Elements of Mechanical Vibration.** By C. R. FREBERG, Ph.D., and E. N. KEMLER, Ph.D. (New York: John Wiley and Sons, Inc.; London: Chapman and Hall, Ltd.) Pp. xiii + 227. Price 22s. 6d. net.

This is the second edition of a book first published in 1943 and now republished with additions which include more work on the vibrations of beams and a chapter on sound. In plan and scope the book is excellent and provides a comprehensive survey of those vibration problems which come within the range of the undergraduate engineering student. Starting with a thorough consideration of vibrations with one degree of freedom it proceeds to systems with a finite number of degrees of freedom and explains the methods of reduction of such systems to a form from which the frequencies of normal modes of vibration may be readily computed. There is a very good chapter on the use of the mobility method, and there are useful accounts of the method of dimensions and of electrical analogies.

From the point of view of the student who wishes to

acquire a working knowledge of these methods the book is good. It would have been very good if the explanations and the treatment of the theoretical aspects of the subject had reached the same standard as the sections dealing with practical applications. In many cases the derivation of results is far from clear, and there are one or two serious errors of principle. In section 7.4 of the chapter on beams, for example, the authors state that the result they obtain for the frequency of vibration of a beam is exact "because the actual deflexion curve is used," whereas they have used the *static* deflexion curve instead of the *dynamic* one.

There are other instances in which lack of clarity and failure to state conditions correctly may prove serious stumbling blocks to the student who tries to follow the authors' reasoning. One point merits special notice and that is the indiscriminate use of the term "weight" to represent either "weight" or "inertia." This appears to be quite purposeless and does not add to the clarity of the explanations.

Perhaps it is only fair to observe that criticisms such as the above could be made of many engineering (and other) textbooks, and that the fact that this book has reached a second edition without the faults being rectified is presumptive evidence that the average reader is interested only in "results" and takes most of the theory for granted.

The book is well produced with clear diagrams and graphs and with many examples worked and unworked, from which the reader should acquire facility in the use of the methods described.

H. T. JESSOP

## Journal of Scientific Instruments

### Contents of the March issue

#### ORIGINAL CONTRIBUTIONS

- An Electrically-maintained Pendulum. By E. B. Brown and A. J. Shimmins.
- A Multi-channel String Galvanometer for use with Wire-resistance Strain Gauges. By A. A. Wells.
- Examination and Calibration of a Babinet Compensator. By H. G. Jerrard.
- A Pendent-Crop Apparatus for Surface and Interfacial Tensions. By H. W. Douglas.
- The Use of a Polarizer in the Disappearing Filament Optical Pyrometer. By R. G. Giovanelli and W. R. G. Kemp.
- Initial Drifts in Running Voltage of Glow-Discharge Regulator Tubes. By F. A. Benson.
- An X-ray Scanning Camera. By J. Thewlis and A. R. Pollock.
- A High-Sensitivity Torsion Creep Unit. By A. E. Johnson.

#### Laboratory and Workshop Notes

- Improvement to a Pantograph. By L. Bainbridge-Bell.
- An Audible Vacuum-leak Indicator. By J. T. Lloyd.
- Hard-soldering Greasy Articles. By G. W. Smith.
- The Electron Gun Filament in the Electron Microscope. By J. Dlugosz.
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## SPECIAL ARTICLE

## The International Temperature Scale—1948 Revision

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The Ninth General Conference of Weights and Measures held in Paris in October 1948, adopted "The International Temperature Scale of 1948," being a revision of "The International Temperature Scale of 1927." This article gives the essential details of the new scale and discusses the changes which have been made and the reasons underlying them. The most important differences are: (1) the use of a value of  $960\cdot8^\circ$  instead of  $960\cdot5^\circ$  C for the freezing point of silver; (2) the adoption of the Planck equation instead of that of Wien for extrapolation to high temperatures; (3) the use of a value of  $1\cdot438$  instead of  $1\cdot432$  for the constant  $C_2$  in the Planck equation; (4) the use of the name "Celsius" instead of "Centigrade." The effect of these changes on the numerical values of temperature is given in graphical form. The reproducibility of the scale is discussed and an estimate is made of the accuracy with which it represents the thermodynamic scale.

The first specification for an International Temperature Scale was authorized by the Seventh General Conference of Weights and Measures in 1927. This was reviewed by the Eighth Conference in 1933, when a few textual errors were corrected but no other modifications were made. An advisory committee (Comité Consultatif de Thermométrie) was then formed to advise future conferences on modifications to the scale so that it might continue to reproduce as closely as possible the thermodynamic Centigrade scale; and, at its meeting in July 1939, it made certain recommendations for changes which should have been submitted to the General Conference in the autumn of that year. Owing to the outbreak of war, the Ninth General Conference did not take place until October 1948, when a revised specification of the scale, known as "The International Temperature Scale of 1948," was adopted in the light of further recommendations from the advisory committee which met in May of that year. This article gives the reasons for making the changes and some details of the supporting evidence. The various page references are to *Procès-Verbaux des Séances du Comité International des Poids et Mesures*, deuxième Série, Vol. 21. The official French text of the scale appears in its final form in *Comptes Rendus des Séances de la Neuvième Conférence Générale des Poids et Mesures*, pp. 89–100, and an English translation is being published by H.M. Stationery Office.

## THE ABSOLUTE SCALE

Before considering the modifications which have been made in the scale, it will be convenient to refer to the consideration given by the advisory committee to the proposal of Giauque that the scale should be defined in terms of the interval between the absolute zero and the ice point. The degree would thus become a prescribed fraction of this interval instead of the interval between the ice and steam points as at present.<sup>(1)</sup> The advantages claimed for such a system are: (i) the relation between temperatures on the absolute thermodynamic scale and

the practical scale is fixed by definition; (ii) temperatures measured with the gas thermometer can be stated more exactly on the proposed scale than on a scale based on the ice and steam points, except in the neighbourhood of these points; (iii) the use of the gas thermometer would become simpler and more convenient, because it would not be necessary to make measurements at the steam point.

It was further proposed that the temperature of the ice point or triple point of water (whichever was chosen as the fundamental point on the scale) should be fixed once and for all. If this were done, and subsequently more precise gas thermometry led to a different value, the size of the degree would be changed, and the numerical values of all temperatures except the ice point and the absolute zero would be changed. Discussion on the value to be adopted for the ice point on the absolute (Kelvin) scale revealed a sharp difference of opinion between the merits of  $273\cdot15^\circ$  and  $273\cdot16^\circ$  K, the former figure being preferred by European workers and the latter by the National Bureau of Standards of the U.S.A. The figure of  $273\cdot15 \pm 0\cdot02^\circ$  K had been recommended by the meeting which took place in July 1939, and it was decided that there was no new evidence to justify making a change.

From this discussion, it was clear that on the Giauque system, the degree could not be defined to better than one part in 27 000, whereas, since the interval between the ice and steam points can be reproduced to an accuracy of about one part in 100 000, the size of the degree of the existing scale is known to nearly four times as great a precision. The practical advantages of this fact to the metrologist weighed strongly with the committee, and, while they gave their approval "in principle" to Giauque's proposal, they were not prepared to recommend its adoption until the value of the ice point on the absolute scale should be more accurately known. Clearly, the numerical value of the interval between the ice and steam points cannot be stable to  $0\cdot001^\circ$  C until the value of the ice point on the Kelvin scale is certain to  $0\cdot0027^\circ$  K.

It is, however, necessary to assume a definite value for the temperature of the ice point on the Kelvin scale for the purpose of calculations involving temperatures on this scale and "for the sake of uniformity" the committee recommended the use of the figure  $273.15^\circ\text{K}$ , but when the proposals of the advisory committee came before the International Committee and the General Conference, this figure was not ratified. An uncertainty of one- or two-hundredths of a degree is hardly significant when the figure is used in Planck's formula (the only purpose for which it is required on the International scale) and the recommendation of the committee that the figure of  $273.15^\circ\text{K}$  be adopted for the sake of uniformity does not seem to be open to objection. This figure is being used by the National Physical Laboratory.

#### NOMENCLATURE

An important change of nomenclature was adopted at the General Conference. In the recommendations of the advisory committee reference was made to "l'échelle thermodynamique centésimale." The word "centésimal" had been recommended by the International Union of Pure and Applied Physics in order to eliminate the ambiguity which otherwise existed in French between the Centigrade temperature scale and the "grade" system of angular measurement which divides the right-angle into a hundred grades. It was not, however, well received by the General Conference and, after some discussion, it was decided to use internationally the name "Celsius" which is already used to describe the scale in many European countries. This alteration does not affect the use of " $^\circ\text{C}$ " as the symbol, it is unambiguous, it brings all countries into line, and there is further consistency in that, by its use, all the common temperature scales are now known by the names of their originators: Celsius, Fahrenheit, Kelvin and Réaumur.

Temperatures measured on the International Temperature Scale of 1948 are to be indicated as  $^\circ\text{C}$  (Int. 1948) wherever it is necessary to indicate that the 1948 scale is in use. The adoption of the word "Celsius" should apply to all scales hitherto known as Centigrade. The change of name is not intended to serve as a means of identifying the 1948 scale.

#### RANGE AND SUB-DIVISION OF THE SCALE

The aim of the International scale is to provide a conveniently and accurately reproducible scale which shall conform as closely as possible, in the light of present knowledge and techniques, with the thermodynamic Celsius scale. To that end, the most probable values on the thermodynamic scale of the fixed points given in Table 1 have been adopted by definition, and specified interpolation instruments are used to realize the scale between them.

This table, taken from the 1948 specification, is, in essence, the same as that adopted in 1927 except that the value of the silver point has been changed from

$960.5$  to  $960.8^\circ\text{C}$ . The reasons for this change are discussed below.

Table 1. *Fixed points on the thermodynamic scale*

<i>Fundamental fixed points and primary fixed points under the standard pressure of 1 013 250 dynes/cm<sup>2</sup></i>	<i>Temperature, <math>^\circ\text{C}</math></i>
(a) Temperature of equilibrium between liquid oxygen and its vapour (boiling point of oxygen)	$-182.970$
(b) Temperature of equilibrium between ice, and air-saturated water (melting point of ice) ( <i>fundamental fixed point</i> )	$0$
(c) Temperature of equilibrium between liquid water and its vapour (boiling point of water) ( <i>fundamental fixed point</i> )	$100$
(d) Temperature of equilibrium between liquid sulphur and its vapour (boiling point of sulphur)	$444.600$
(e) Temperature of equilibrium between solid and liquid silver (freezing point of silver)	$960.8$
(f) Temperature of equilibrium between solid and liquid gold (freezing point of gold)	$1\,063.0$

According to the 1948 specification, interpolation between these fixed points is to be carried out as follows:

(a) From  $0^\circ\text{C}$  to the freezing point of antimony the temperature  $t$  is defined by the formula

$$R_t = R_0(1 + At + Bt^2)^*$$

where  $R_t$  is the resistance, at temperature  $t$ , of the platinum wire between the branch points formed by the junctions of the current and potential leads of a standard resistance thermometer. The constant  $R_0$  is the resistance at  $0^\circ\text{C}$ , and the constants  $A$  and  $B$  are to be determined from measured values of  $R_t$  at the boiling points of water and sulphur. The platinum in a standard resistance thermometer shall be annealed and of such purity that  $R_{100}/R_0$  is greater than  $1.3910$ .

(b) From the boiling point of oxygen to  $0^\circ\text{C}$ , the temperature  $t$  is defined by the formula

$$R_t = R_0[1 + At + Bt^2 + C(t - 100)t^3]^\dagger$$

where  $R_t$ ,  $R_0$ ,  $A$  and  $B$  are determined in the same manner as in (a) above and the constant  $C$  is calculated from the measured value of  $R_t$  at the boiling point of oxygen.

(c) From the freezing point of antimony to the freezing point of gold the temperature  $t$  is defined by the formula

$$E = a + bt + ct^2$$

where  $E$  is the electromotive force of a standard thermocouple of platinum and rhodium-platinum alloy, when one junction is at  $0^\circ\text{C}$  and the other is at the temperature  $t$ . The constants  $a$ ,  $b$ , and  $c$  are to be calculated from measured values of  $E$  at the freezing points of antimony, silver and gold. The antimony used in

\* Equivalent to the Callendar formula

$$t - t_{pt} = \delta(t - 100)t10^{-4}$$

† Equivalent to the Callendar-van Dusen formula

$$t - t_{pt} = \delta(t - 100)t10^{-4} + \beta(t - 100)t310^{-8}$$



determining these constants shall be such that its freezing temperature, determined with a standard resistance thermometer, is not lower than  $630.3^{\circ}\text{C}$ . Alternatively, the thermocouple may be calibrated by direct comparison with a standard resistance thermometer in a bath at any uniform temperature between  $630.3$  and  $630.7^{\circ}\text{C}$ .

The platinum wire of the standard thermocouple shall be annealed and of such purity that the ratio  $R_{100}/R_0$  is greater than  $1.3910$ . The alloy wire shall consist nominally of 90% platinum and 10% rhodium by weight. When one junction is at  $0^{\circ}\text{C}$  and the other at the freezing point of antimony ( $630.5^{\circ}\text{C}$ ), silver or gold, the completed thermocouple shall have electromotive forces, in microvolts, such that

$$E_{Au} = 10\,300 \pm 50 \mu\text{V}$$

$$E_{Au} - E_{Ag} = 1\,185 + 0.158(E_{Au} - 10\,310) \pm 3 \mu\text{V}$$

$$E_{Au} - E_{Sb} = 4\,776 + 0.631(E_{Au} - 10\,310) \pm 5 \mu\text{V}$$

(d) Above the gold point the temperature  $t$  is defined by the formula

$$\frac{J_t}{J_{Au}} = \frac{\exp [c_2/\lambda(t_{Au} + T_0)] - 1}{\exp [c_2/\lambda(t + T_0)] - 1}$$

in which  $J_t$  and  $J_{Au}$  are the radiant energies per unit wavelength interval at wavelength  $\lambda$ , emitted per unit time by unit area of a black body at the temperature  $t$  and at the freezing point of gold,  $t_{Au}$ , respectively.

$c_2$  is  $1.438$  cm degrees.

$T_0$  is the temperature of the ice point in  $^{\circ}\text{K}$ .

$\lambda$  is a wavelength of the visible spectrum.

This part of the text differs in several important particulars from that adopted in 1927. Originally, the resistance thermometer, calibrated at the boiling point of oxygen ( $-182.97^{\circ}\text{C}$ ) was used to define the scale down to  $-190^{\circ}\text{C}$  by extrapolation. It has been found, however, that the temperatures so deduced in the range  $-182.97$  to  $-190^{\circ}\text{C}$  were too high according to the gas thermometer, the difference rising to  $0.03$  or  $0.04$  at  $-190^{\circ}\text{C}$ . In consequence, the international scale is not now carried below the boiling point of oxygen. At high temperatures the upper limit of the scale, formerly imposed by the use of Wien's Law in the radiation scale, has been removed by the adoption of Planck's Law.

The link between the parts of the scale defined by the resistance thermometer and the platinum/10% rhodium-platinum thermocouple is provided by the freezing point of a particular sample of antimony as determined by the platinum resistance thermometer. In the 1948 specification, as an alternative, a direct comparison between the resistance thermometer and thermocouple may be made between  $630.3^{\circ}\text{C}$  and  $630.7^{\circ}\text{C}$  in a suitable comparison bath. In the 1927 scale, though the thermocouple and resistance thermometer were made to agree at the antimony point the scale was defined by the resistance thermometer up to  $660^{\circ}\text{C}$ . It was subsequently found

that the scales defined by the two instruments diverged in the range  $630.5^{\circ}$  to  $660^{\circ}\text{C}$ , the difference amounting to one- or two-tenths of a degree at  $660^{\circ}\text{C}$ . Therefore, in the 1948 scale, the change from one interpolation instrument to the other, is made at the antimony point itself.

#### THE ZERO POINT

Opinion during recent years has been that the triple-point of water would provide a more reproducible fixed point than the ice point. The values for the triple point obtained by different observers range from  $0.0098^{\circ}$  to  $0.00993^{\circ}\text{C}$ , and it would obviously be inconvenient to change the zero point of the scale by about a hundredth of a degree. It will be noted from the table of fixed points specified in the scale that the ice and steam points are singled out as "fundamental" instead of "primary" and are distinguished by having no figures given after the decimal point. It therefore seemed desirable to keep  $0^{\circ}\text{C}$  as one of the fundamental points and the ice point has been retained as the zero point of the scale. It is now recommended, however, that for the most accurate work it should not be reproduced directly, but by the use of the triple point, to which a value of  $0.0100^{\circ}\text{C}$  has been assigned. This figure is just outside the range of observed values of the triple point, but as it is doubtful whether the melting point of ice can ever be determined to a greater accuracy than  $\pm 0.0001^{\circ}\text{C}$ , owing to the difficulty of reproducing the condition of "water saturated with air at  $0^{\circ}\text{C}$ ," a discrepancy of one in the fourth decimal place is hardly significant.

The triple point is easily reproduced in a cell fitted with a re-entrant tube for the thermometer, and containing nothing but pure water and its vapour. Using such cells, several workers have been able to maintain a temperature constant to within  $0.0001^{\circ}\text{C}$  over a period of days.<sup>(2, 3, 4, 5)</sup> The difficulties in the melting-point determinations arising from contamination of the ice-water mixture and the effects of dissolved carbon dioxide are eliminated. The difference between the values of the two points results from the difference in pressure (about  $756$  mm of mercury) and the absence of depression due to the presence of dissolved gases. The pressure effect accounts for about  $0.0075^{\circ}$  and the dissolved gases for about  $0.0025^{\circ}\text{C}$ .

#### THE FREEZING-POINT OF SILVER

The discrepancy already mentioned between the temperatures defined by the thermocouple and the resistance thermometer in the range  $630.5^{\circ}$  to  $660^{\circ}\text{C}$  showed that the transition from one part of the scale to the other was not perfectly smooth. In order to eliminate this defect, the National Bureau of Standards proposed that the numerical value of the silver point should be changed from  $960.5^{\circ}\text{C}$  to  $960.8^{\circ}\text{C}$ , a change which would not carry the value outside the range of experimental error in the gas thermometer determinations of the point. In some unpublished work on the ratio of the intensities

from black bodies immersed in freezing silver and freezing gold, they had also found a value of  $960.8^{\circ}\text{C}$  when  $1.063^{\circ}\text{C}$  was taken as the gold point and  $1.438\text{ cm deg.}$  as the constant  $c_2$  in the radiation formula.

Investigations carried out at the National Physical Laboratory showed (pp. T73-4)\* that the difference between the temperature as measured by the resistance thermometer and by the thermocouple over the range  $550^{\circ}$  to  $630^{\circ}\text{C}$  is at a minimum when the silver point is fixed at a value between  $960.8^{\circ}$  and  $960.9^{\circ}\text{C}$  (Fig. 1).

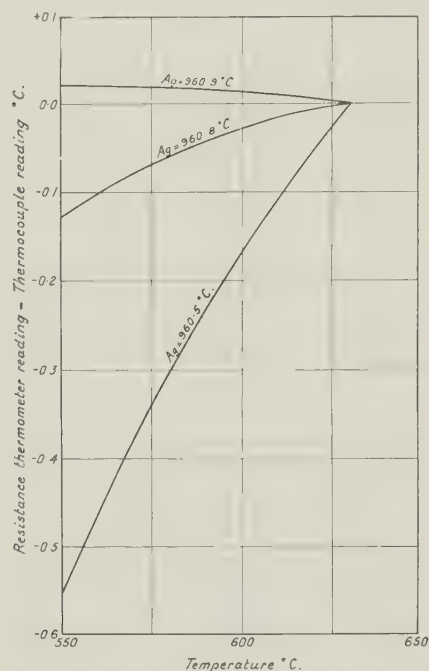


Fig. 1. Difference between resistance thermometer and thermocouple readings on the basis of different silver point values

At the higher temperatures subsequent work has tended to confirm the Bureau of Standards' conclusions regarding the overlap with the radiation scale. The weight of evidence thus seemed in favour of the value  $960.8^{\circ}\text{C}$ , and this figure was therefore adopted.

#### THE RADIATION CONSTANT, $c_2$

The value of the radiation constant,  $c_2$ , has been changed from  $1.432\text{ cm deg.}$  The higher value of  $c_2$  is based principally on the critical examination by Birge of the values of the atomic constants leading to  $1.43848$ .<sup>(6)</sup> Dumond and Cohen of the California Institute of Technology, in an unpublished work, have arrived at the value  $1.43847 \pm 0.00019$  by using Birge's data and incorporating the results of new experiments. Radiation measurements tend to give a somewhat lower value although the recent work at the National Bureau of Standards by Van Dusen and Dahl gives a mean

value of  $1.4382$ .<sup>(7)</sup> These measurements were of the ratio of intensity of the radiation from a black body at the freezing points of cobalt and nickel to that of the freezing point of gold, and these have been combined with an older measurement by Fairchild and Hoover for palladium. The ratios were determined by means of the disappearing filament optical pyrometer. The metal used in each case was either from the batch of metal employed by Day and Sosman in their gas thermometer determinations of the freezing points or could be correlated with it, thus the gas thermometer temperature could be used and the corresponding value of  $c_2$  calculated. The values ranged from  $1.436_4$  to  $1.439_6$ .

#### THE STANDARD RESISTANCE THERMOMETER AND THE STANDARD THERMOCOUPLE

In addition to the changes in the fundamental constants of the scale which have already been considered, changes were made in the specifications of the standard platinum resistance thermometer and the standard platinum/10% rhodium-platinum thermocouple. In 1927 the purity of the platinum to be used in either instrument was specified by means of the ratio  $R_{100}/R_0$ , which was laid down as being not less than  $1.390$ . This figure has now been increased to  $1.391$ .

The only control over the alloy wire of the thermocouple in the 1927 specification was that the e.m.f. of the thermocouple at the gold point should lie between  $10.200$  and  $10.400$  microvolts. This tolerance is needlessly wide, and in the 1948 specification it has been reduced to  $10.300 \pm 50\text{ }\mu\text{V}$ . This specification alone, however, gives no indication that the alloy wire consists of pure platinum and pure rhodium, and a study of the subject at the National Physical Laboratory led to the conclusion that impurities present in the alloy wire could lead to perceptible changes in curvature of the e.m.f.-temperature curve (pp. T73-82).

The results of a large number of calibrations of thermocouples of American, British and German origin were analysed, and it was found that by plotting the differences between the e.m.f.'s at the gold and antimony points ( $E_{Au} - E_{Sb}$ ) and the gold and silver points ( $E_{Au} - E_{Ag}$ ) against the e.m.f.'s at the gold point, two straight lines were obtained, such that the following relationships held:

$$E_{Au} - E_{Ag} = 1.185 + 0.158 (E_{Au} - 10.310) \pm 3\text{ }\mu\text{V} \quad \text{absolute electrical units}$$

$$E_{Au} - E_{Sb} = 4.476 + 0.631 (E_{Au} - 10.310) \pm 5\text{ }\mu\text{V} \quad \text{absolute electrical units}$$

It was shown that thermocouples which just failed to satisfy these criteria gave a calibration curve which was just measurably different from the normal in shape and the wires were found to be just perceptibly less pure than the others when examined spectroscopically. In view of this evidence the equations have been adopted as obligatory criteria for the purity of standard couples to be used for defining the scale.

\* See opening paragraph of this article.



# THE EFFECT OF THE CHANGES IN THE SCALE ON THE NUMERICAL VALUES OF TEMPERATURE

In general, the changes in the scale will only be appreciable in work of the highest precision. From  $-183^{\circ}$  to  $630.5^{\circ}\text{C}$  the scale, in fact, remains unaltered except that it should now be slightly more clearly defined. Over the range from  $630.5^{\circ}$  to  $1063^{\circ}\text{C}$  the nominal value of any temperature is increased owing to the change in value of the silver point from  $960.5^{\circ}$  to  $960.8^{\circ}\text{C}$ , the maximum difference of  $0.4^{\circ}\text{C}$  occurring at about  $50^{\circ}\text{C}$ . Above  $1063^{\circ}\text{C}$  the numerical values of all temperatures are decreased by progressively greater amounts as the temperature increases. This change is mainly accounted for by the change in  $c_2$  from  $1.432$  to  $1.438$ , but other small differences are due to the use of the value of  $273.15^{\circ}$  for conversion to degrees Kelvin instead of  $273^{\circ}$  as in the 1927 scale, and above about  $2500^{\circ}\text{C}$  to the use of the Planck formula instead of that of Wien. The new values of the secondary fixed points given in Table 2 reflect these changes.

Table 2. *New values of secondary fixed points*

Secondary fixed points under the pressure of one standard atmosphere (except for the triple points)	Temperature $^{\circ}\text{C}$ (Int. 1948)
Temperature of equilibrium between solid carbon dioxide and its vapour	$-78.5$
Temperature of freezing mercury	$-38.87$
Temperature of equilibrium between ice, water and its vapour (triple point)	$0.0100$
Temperature of transition of sodium sulphate decahydrate	$32.38$
Temperature of triple point of benzoic acid	$122.36$
Temperature of equilibrium between naphthalene and its vapour	$218.0$

$$t_p = -78.5 + 12.12\left(\frac{p}{p_0} - 1\right) - 6.4\left(\frac{p}{p_0} - 1\right)^2$$

Temperature  $^{\circ}\text{C}$  (Int. 1948)

$-78.5$

$-38.87$

$0.0100$

$32.38$

$122.36$

$218.0$

$$t_p = 218.0 + 44.4\left(\frac{p}{p_0} - 1\right) - 19\left(\frac{p}{p_0} - 1\right)^2$$

\* These equations are intended for use over the range from  $p = 680$  to  $p = 780$  mm of mercury.

Table 2. *New values of secondary fixed points* (continued)

Secondary fixed points under the pressure of one standard atmosphere (except for the triple points)	Temperature $^{\circ}\text{C}$ (Int. 1948)
Temperature of freezing tin	$231.9$
Temperature of equilibrium between benzo-phenone and its vapour	$305.9$
$t_p = 305.9 + 48.8\left(\frac{p}{p_0} - 1\right) - 21\left(\frac{p}{p_0} - 1\right)^2$	
Temperature of freezing cadmium	$320.9$
Temperature of freezing lead	$327.3$
Temperature of equilibrium between mercury and its vapour	$356.58$
$t_p = 356.58 + 55.552\left(\frac{p}{p_0} - 1\right) - 23.03\left(\frac{p}{p_0} - 1\right)^2 + 14.0\left(\frac{p}{p_0} - 1\right)^3$	
Temperature of freezing zinc	$419.5$
Temperature of freezing antimony	$630.5$
Temperature of freezing aluminium	$660.1$
Temperature of freezing copper in a reducing atmosphere	$1083$
Temperature of freezing nickel	$1453$
Temperature of freezing cobalt	$1492$
Temperature of freezing palladium	$1552$
Temperature of freezing platinum	$1769$
Temperature of freezing rhodium	$1960$
Temperature of freezing iridium	$2443$
Temperature of melting tungsten	$3380$

These values have all been derived by calculation from those suggested in 1927 and 1939, and do not represent new determinations of the points. The precise differences between the 1927 and 1948 scales throughout the range  $630^{\circ}$  to  $5000^{\circ}\text{C}$  are shown in Fig. 2.

## ACCURACY OF THE SCALE

Since, in general, the interpolation instruments used in the realization of the international scale are capable of

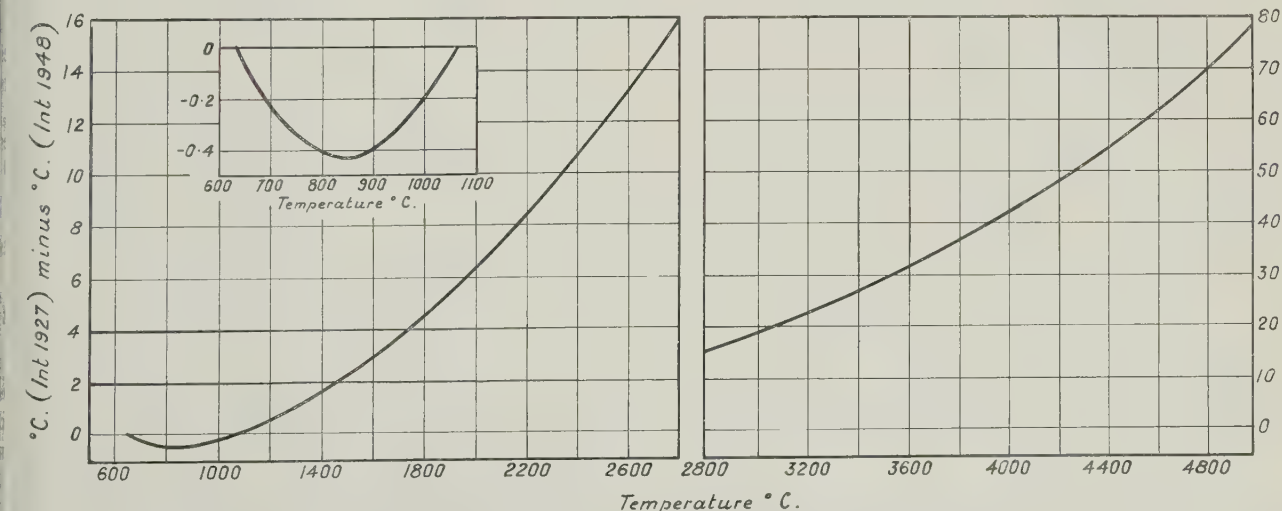


Fig. 2. Changes in numerical value of temperature resulting from change from "The International Temperature Scale of 1927" to "The International Temperature Scale of 1948"

a reproducibility of reading greater than the accuracy of the fundamental instruments by which the thermodynamic scale is realized, it is necessary to draw a distinction between accuracy and reproducibility. For example, the value of the gold point is taken as  $1\,063^\circ\text{C}$  on the basis of two gas-thermometer determinations giving values of  $1\,062\cdot4^{(8)}$  and  $1\,063\cdot5^\circ\text{C}$ ,<sup>(9)</sup> of which the former is the more recent. Clearly, the value on the thermodynamic scale cannot be known to better than  $\pm 0\cdot5$  or  $\pm 1^\circ\text{C}$ , but the point can be readily reproduced to  $\pm 0\cdot1^\circ\text{C}$  when the platinum thermocouple is used as the measuring instrument.

The curves of Fig. 3 give a rough guide to the degree of reproducibility of the international scale. At the zero point it is much higher than elsewhere, since it depends solely on the reproducibility of the triple point of water, and is independent of any possible differences between the calibrations of a number of resistance thermometers. The absolute accuracy of this point on the thermodynamic scale, however, depends on the precision with which the difference between the ice point and the triple point is known. This is taken as  $0\cdot0100^\circ\text{C}$ , but might, on available experimental evidence, be as low as  $0\cdot0098^\circ\text{C}$ ; there is thus an uncertainty in one direction of about  $0\cdot0002^\circ\text{C}$ . At the steam point, there is no distinction between accuracy and reproducibility, since each is determined solely by the precision with which the boiling point may be realized, about  $\pm 0\cdot001^\circ\text{C}$ .

The boiling point of oxygen may be reproduced to about  $\pm 0\cdot002^\circ\text{C}$ , but the value on the thermodynamic scale is not known to better than about  $\pm 0\cdot02^\circ\text{C}$ . The difference between the thermodynamic and international scales of temperature in the range from  $0^\circ$  to the boiling point of oxygen is less than  $0\cdot05^\circ$ , but there is not agreement on the sign of some of the differences which are of the same order of magnitude as the uncertainties in gas-thermometer measurements.

At the sulphur boiling point, reproducibility is of the order of  $\pm 0\cdot002^\circ\text{C}$ , but there is appreciably greater uncertainty in the absolute accuracy. The international scale assumes a value of  $444\cdot600^\circ\text{C}$ , which is in agreement to within  $\pm 0\cdot1^\circ\text{C}$  of nearly all the gas thermometer determinations of the point. Recently, however, workers at the Massachusetts Institute of Technology have obtained a value of  $444\cdot74^\circ\text{C}$ .<sup>(10)</sup> On the basis of this value they have given a formula to express the differences between the international and thermodynamic scales over the range  $100^\circ$  to  $444\cdot74^\circ\text{C}$ . This formula is given in Part IV (Supplementary Information) of the text of the "International Temperature Scale," and leads to differences of  $0\cdot05^\circ$  at  $200^\circ\text{C}$  and  $0\cdot12^\circ$  at  $300^\circ\text{C}$ . The evidence, however, was not thought to be sufficiently strong to justify changing the value of the sulphur point from  $444\cdot600^\circ\text{C}$ .

The uncertainty, therefore, in the agreement between the thermodynamic and international scales is of the

order of  $\pm 0\cdot1^\circ\text{C}$  at the sulphur point and rises to nearly  $\pm 1^\circ\text{C}$  at the gold point. There is little direct evidence as to the agreement between the scales in the intervening range of temperature, but extrapolation of the resistance thermometer law up to the gold point leads to a value only about a degree below the accepted figure and a smooth transition from the resistance thermometer scale to the thermocouple scale at  $630^\circ\text{C}$  may be achieved with a reasonable value for the silver point. It thus seems likely that the divergence between the two scales is not much greater than would be obtained by assuming a progressive increase from  $\pm 0\cdot1^\circ$  at sulphur to  $\pm 1^\circ$  at gold.

At the upper end of the scale shown in Fig. 3 ( $2\,000^\circ\text{C}$ )

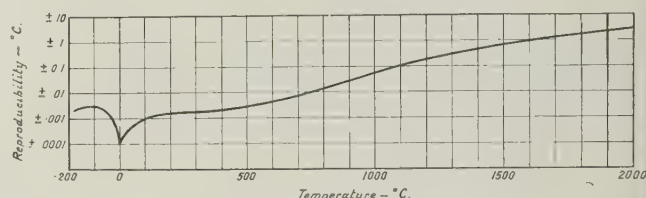


Fig. 3. Reproducibility of International Temperature Scale

the reproducibility takes into account such factors as the accuracy of adjustment of the pyrometer-lamp filament to disappearance, the possible errors in the sectors used for extrapolation and the errors in the determination of the effective wavelength of the pyrometer. Assuming the validity of Planck's law, the uncertainty in the knowledge of the thermodynamic scale at this temperature is made up of two parts: an uncertainty of  $\pm 1^\circ\text{C}$  in the gas-thermometer value of the gold point is equivalent to  $\pm 3^\circ\text{C}$  at  $2\,000^\circ\text{C}$ , and if we admit an uncertainty of  $\pm 0\cdot003$  in  $c_2$  on account of the discrepancy between the values found by direct radiation measurements and those deduced from atomic constants, we must add a further  $3^\circ\text{C}$ .

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## ORIGINAL CONTRIBUTIONS

## Measuring a Large Displacement by Interferometry

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A modified form of Michelson interferometer is used to calibrate the mechanical displacement system of an adjustable cavity resonator. With this resonator the velocity of propagation of electromagnetic waves has been newly determined, as will be described elsewhere. For the calibration a linear displacement of 12.7 cm (5 in) was determined in standard krypton wavelengths to an accuracy of one part in a million, utilizing techniques of fringe observation and calculation which do not involve counting a progression of fringes. Displacements up to 30 cm and greater can be similarly measured if suitable sources of monochromatic radiations are employed.

Interference methods of measuring linear displacements are mostly applied when the displacements are small and permit observation of the progression of interference fringes. An exceptional application was made by Michelson and Benoît when measuring a mirror displacement of about 0.39 mm by counting the passage of 1212 fringes during the original determination of the metre in waves of cadmium red light. If it were necessary, such a task would now be accomplished with the aid of a cine-camera or a photoelectric counter. It is not commonly realized, however, that modern interferometers and sources of monochromatic light can be employed for measuring displacements of 30 cm or greater without the need for counting fringes. The following account of the measurement of a 12.7 cm (5 in) displacement to an accuracy of one part in a million illustrates the present potentialities of interferometry.

The work described was undertaken to calibrate the displacement system in an adjustable cylindrical cavity resonator. This resonator has been used in the Electricity Division of the N.P.L. for new determinations of the velocity of propagation of electromagnetic waves. In the earlier determinations made by Essen and Gordon-Smith,<sup>(1)</sup> a resonator of fixed length was used. The present resonator is fitted with a piston for varying the cavity length, the purpose being to eliminate certain possibilities of systematic error from the new results. Displacements of the piston are controlled by means of end-standards, known to the precision engineer as slip or block gauges. The precise relationship between these displacements and the actual amounts of travel of the piston head along the axis of the resonator was determined by interferometry. The work was entirely subsidiary to the velocity determinations, but is believed to be of sufficient metrological interest to warrant separate description.

## THE OPTICAL AND DISPLACEMENT SYSTEMS

The Michelson interferometer is well adapted to the measurement of displacement. A distinguishing characteristic of its optical system is the division of the incident beam into two parts which advance and return, after reflexion at mirrors, along directions usually at right

angles to one another. If one mirror is displaceable and the other fixed at approximately the same distance from the beam divider as the mean position of the movable mirror, the maximum difference of path length involved in a displacement  $L$  is  $2L/2$ , compared with  $2L$  for the same displacement in the Fizeau and Fabry-Perot interferometers. This feature has proved attractive to designers of special instruments for length measurement.

One such instrument is the Kösters gauge interferometer<sup>(2)</sup> used in standards laboratories for calibrating slip and block gauges. It employs the Michelson principle in the modified form introduced by Twyman and Green.<sup>(3)</sup> The mirrors are optically inclined to one another at a small angle and are illuminated with parallel monochromatic light, so that fringes of constant thickness are observed. A constant-deviation prism is incorporated for illuminating the field in turn with different radiations from the one source, such as a cadmium or a krypton lamp. From the fringe observations made in various radiations, the path difference between the interfering beams is derived by the method of excess fractions due to Benoît and Michelson.<sup>(4)</sup>

The general arrangement of the apparatus is shown

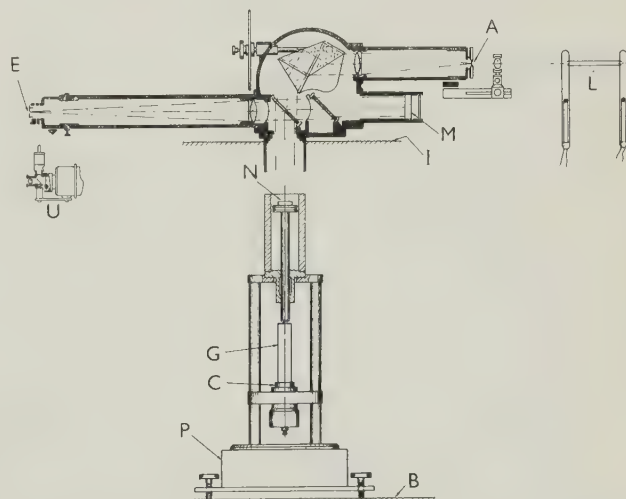


Fig. 1. General arrangement of optical and displacement systems

in Fig. 1 and certain features of the Kösters instrument,<sup>(2)</sup> will be recognized in the upper part. Light from a hot-cathode krypton lamp  $L$  (by The General Electric Co., Ltd.) is condensed on a small rectangular aperture  $A$  ( $0.04 \times 0.3$  mm) situated at the principal focus of a collimating lens of 21 cm focal length. By suitable adjustment of the micrometer head controlling rotation of the constant-deviation prism, a parallel beam originating from a single krypton line is directed on to the beam divider. Here the beam is separated into two parts of approximately equal intensity, one travelling horizontally to the fixed reference mirror  $M$  through the inclined compensator plate as in the normal Michelson system. The other falls vertically on to the second interferometer mirror  $N$ , constituted by the surface of a thin slip gauge which is attached by soft wax to the piston head. Light reflected from the piston mirror and the reference mirror is partly recombined at the beam divider and the resultant beam is viewed directly through the small exit aperture  $E$  ( $0.15 \times 1.5$  mm) situated at the principal focus of the objective lens. This lens has a focal length of 42 cm.

The Kösters interferometer is fitted with an auto-collimating eyepiece which is used solely for checking the optical adjustments. For this purpose the eyepiece unit  $U$  is swivelled into position behind the exit aperture, on which it directs light from a small electric lamp. The aperture is adjusted in its holder until the aperture and its real image viewed in light reflected from the reference mirror are coincident. This ensures that the optical axis of the interferometer, defined by the centres of the aperture and the objective lens, is normal to the reference mirror. Similar use is then made of the eyepiece to adjust the piston mirror. If this mirror happens to be so maladjusted that the reflected image cannot be seen through the aperture, a graticule ruled on glass is temporarily substituted for the aperture holder so that a larger field is available for preliminary adjustment. Both adjustments are essential for assuring that the optical measurements are free from any appreciable obliquity error due to lack of parallelism between the plane wave fronts and the mirrors.

The whole apparatus was arranged so that the average length of path traversed by the two interfering beams could be approximately equalized. The optical head was rigidly attached, in an overhanging position, to the slate top  $I$  of a massive, isolated concrete pillar about 1 m above floor level. The resonator was placed underneath and was clamped over a packing block  $P$  to a levelling table resting on a ledge  $B$  of the same pillar at floor level. The flexibility of adjustment provided by use of the packing block and levelling table enabled the desired relationship between the mirrors to be satisfactorily achieved.

The case-hardened mild steel cylinder of the resonator possesses a highly finished bore of about 6.5 cm diameter and 18 cm long. As designed for the velocity determinations, it is supported with its axis vertical in a

tripod over the centre of a lapped, hardened steel base plate  $C$  of 4 cm diameter. A plunger, extending downwards from the piston head, passes through a good slide-fitting plain bearing at the bottom of the cylinder and is provided with a steel ball tip at its lower end. Controlled piston displacements are made by wringing slip gauges  $G$  of known size in turn to the base plate and bringing the ball tip successively into contact with the upper surface of each gauge. The piston plunger is attached to a counterbalancing system comprised of two weights, suspended with flexible wires over pulleys, which are adjusted so that a small downward force acts on the plunger. If the surface of the base plate is not square to the cylinder axis, a displacement of  $L$  at the ball tip in a direction normal to the base plate surface produces a piston movement of  $L \sec \theta$  along the cylinder axis,  $\theta$  being the angular error of squareness.

The fringes become visible when the levelling table under the resonator is appropriately adjusted. They appear to be located over the superposed surfaces of the reference and piston mirrors,  $M$ ,  $N$ , both of which appear in sharp focus in the field. The separation and orientation of the fringes is under control of the levelling table adjustments.

Before fringe observations were made, the piston mirror  $N$  was adjusted into "squareness" with the cylinder axis. A test was made by rotating the piston through half a turn and noting the change in the number of fringe spaces superposed over the longer dimension (3.5 cm) of the piston mirror. Using the soft wax attachment of the mirror as the medium of adjustment, the change during rotation was finally reduced to about 6 fringe spaces in green light, corresponding to a "squareness" error within 5 sec of arc

$$(\cos 5'' = 1 - 3 \times 10^{-10} \text{ approximately}).$$

Appropriate readjustments of the resonator levelling table were made after each adjustment of the mirror in order to return to the correct relationship with the optical head.

For electrical reasons the piston head is constructed so that an annular gap exists between its cylindrical surface and the resonator wall. The fit of the plain bearing which guides the piston is designed to allow reasonably easy motion of the plunger and yet prevent the gap at the head from becoming appreciably eccentric. Tests made by interferometry showed that an angular displacement of the piston head amounting to 25 seconds of arc could be obtained by forcible manipulation of the plunger. If the plunger was allowed to attain its rest positions by downward displacement under gravity, the variations in final angular position rarely exceeded 1 second of arc.

#### OBSERVATION AND CALCULATION OF DISPLACEMENT

The amount of piston mirror displacement is derived from two sets of observations of fringe position in the



field with respect to a fixed datum point. One set is made before and the other after the piston is displaced, each set consisting of readings obtained when the field is illuminated with the seven krypton radiations in turn.

Fig. 2 represents a typical view of the interferometer

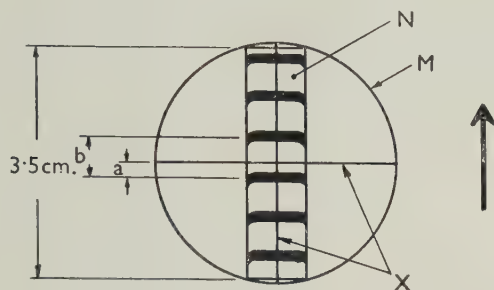


Fig. 2. Diagram of the interferometer field of view

field in monochromatic light. On the reference mirror *M*, which is a glass optical flat fully aluminized on its front surface, are scribed two graticule lines, *X*, the intersection of which constitutes the datum point in the field. The graticule is rendered visible by dimly illuminating the mirror through its back surface with white light.

A simple test is made, while slowly displacing the piston downwards, i.e. away from the beam divider, to ascertain in which direction in the field the order numbers of the fringes increase. Assuming that this direction is upwards in Fig. 2, a visual estimation is made of the distance *a* of the datum point from the lower adjacent fringe, expressed as a fraction of the fringe spacing *b*. Fractional estimations are made, to the nearest 0.05, in the seven krypton lines in sequence, the estimations being then repeated in reverse sequence and the mean values derived.

Let the seven excess fractions thus obtained be represented by  $f'_r$ ,  $r = 1, 2, \dots, 7$ , when the plunger rests on the shorter slip gauge of length  $G_1$ . Also let the corresponding mean fractions be  $f''_r$  when the plunger rests on the longer slip gauge of length  $G_2$ . The seven differences  $(f'_r - f''_r)$  are the fractional parts  $f_r$  of the orders of interference corresponding to the piston mirror displacement  $L \sec \theta$ ,  $L$  being equal to  $(G_2 - G_1)$ .

The Benoît-Michelson method could now be directly applied to determine the integral parts  $N_r$  of the orders of interference in the following equations:—

$$L \sec \theta = (N_r + f_r) \frac{\lambda_r}{2} \quad r = 1, 2, \dots, 7$$

In practice, a convenient modification of the method is used.<sup>(2)</sup> For this, the excess fractions  $F_r$ ,  $r = 1, 2, \dots, 7$ , calculated from the nominal value  $G$  of  $(G_2 - G_1)$ , are subtracted from the corresponding values of  $f_r$  to form the series of difference fractions  $(f_r - F_r)$ . These fractions relate to the small difference  $(L \sec \theta - G)$  and are accordingly associated with small integers. The Benoît-Michelson method becomes much less laborious to apply. Alternatively, a special slide

rule due to Kösters<sup>(2)</sup> may be used to determine the value of  $(L \sec \theta - G)$  directly in metric or inch units from the difference fractions.

Information on the wavelengths and associated data required for displacement measurement is given in Table 1. The values given in the second column repre-

Table 1. Wavelength data for seven krypton lines

Line	Wavelength ( $\mu$ )	Wavelength ratios*	Half-wavelength $\lambda/2$ ( $10^{-6}$ in)	Number of $\lambda/2$ in 5 in
1	0.645 632 25	—	12.709 318 3	393 412.13
2	0.605 615 77	1.066 075 69	11.921 590 9	419 407.11
3	0.599 388 09	1.077 152 28	11.798 998 6	423 764.78
4	0.587 094 57	1.099 707 41	11.556 999 8	432 638.24
5	0.564 959 07	1.142 794 73	11.121 260 8	449 589.31
6	0.557 031 76	1.159 058 24	10.965 211 1	455 987.57
7	0.450 237 80	1.433 980 55	8.862 964 2	564 145.35

\* For use with the modified Benoît-Michelson method, these ratios are rounded to 4 or 5 significant figures.

sent the wavelengths in air, at 20° C and 760 mm pressure, containing water vapour at 7 mm pressure and the normal amount of  $\text{CO}_2$  (0.03% by volume). These have been determined at the N.P.L. from interferometric comparisons with the international reference wavelength of the cadmium red line, and are expressed as decimal fractions of  $1 \mu$ . The wavelength ratios  $\lambda_1/\lambda_r$ ,  $r = 2, 3, \dots, 7$ , given in the third column are for use in the Benoît-Michelson method, and so also are the numbers of half-wavelengths in 5 in. quoted in the fifth column. The latter are derived from the half-wavelengths expressed in micro-inches, the conversion equation being  $1 \mu = 39.370 147 \times 10^{-6}$  in.

#### EXPERIMENTAL PROCEDURE

Although the apparatus was set up in a constant temperature room and well screened, temperature rises generally occurred because of the presence of observers. To compensate for the effects of any uniform temperature drift, every set of fringe position observations in the seven krypton lines was repeated in reverse order, as previously mentioned. Furthermore, if the observations related first to an upward displacement, the whole sequence was repeated in reverse order for a downward displacement, or *vice versa*. By symmetrically relating the fringe observations, and the readings of temperature, atmospheric pressure and humidity, to the mean time of the double sequence, the effects of temperature drift, if reasonably uniform, were eliminated from the results.

In the velocity determinations a set of high quality slip gauges in inch and decimal inch sizes was used. The maximum displacement employed was of the order of 12.7 cm (5 in), controlled by slip gauges of approximately 0.5 in and 5.5 in sizes. For calibration of the displacement system, laboratory standards of 0.5 in and 5.5 in sizes were used which, like the set just mentioned, had been calibrated by interferometry. Interchanges of the gauges were made as rapidly as possible with the gloved hands. The temperature of each

Table 2. *Typical Set of Observations and Calculations*

		Excess fractions in 7 krypton lines. Unit = 0.01 λ/2							
Gauge and temperature		1	2	3	4	5	6	7	D*
G <sub>2</sub> 19.617° C	f''	40	80	90	45	90	00	40	
		65	90	80	55	80	00	40	
	Means	52	85	85	50	85	00	40	
		55	80	45	80	00	50	80	
G <sub>1</sub> 19.604° C	f'	50	80	40	75	00	50	70	
	Means	52	80	42	77	00	50	75	
	f	00	95	57	27	15	50	35	
	F	13	11	78	24	31	57	35	
	(f - F)	- 13	- 16	- 21	+ 03	- 16	- 07	- 00	
	Order	- 0.09	- 0.10	- 0.10	- 0.10	- 0.10	- 0.10	- 0.13	- 1.1
G <sub>1</sub> 19.622° C	f'	90	00	90	00	20	80	70	
		80	90	70	90	05	75	60	
	Means	85	95	80	95	12	77	65	
		70	50	60	05	05	30	20	
G <sub>2</sub> 19.658° C	f''	80	70	65	15	10	35	30	
	Means	75	60	62	10	07	32	25	
	f	10	35	18	85	05	45	40	
	F	13	11	78	24	31	57	35	
	(f - F)	97	24	40	61	74	88	05	
	Order	5.00	5.33	5.38	5.50	5.71	5.79	7.17	+ 63.5
Mean D (Uncorrected)									+ 31.2
Wavelength correction factor									
Air temperature	..	20.08° C	} - 1.18 × 10 <sup>-6</sup>						- 5.9
Barometric pressure	..	763.76 mm							
Vapour pressure	..	8.6 mm							
Mean D (Corrected)									+ 25.3

*Comparison of displacements*

$G_2$ at 19.637° C	=	5.500 011 in
$G_1$ at 19.613° C	=	0.499 999

$L = G_2 - G_1$	=	5.000 012
$L \sec \theta$	=	5.000 025

$$\therefore L(\sec \theta - 1) = +0.000 013$$

\* Piston mirror displacement expressed as the error from 5.0 in. in micro-inches.

gauge during use was related by a sensitive thermocouple to the reading of a standard mercury-in-glass thermometer calibrated to  $0.002^\circ \text{C}$ . One joint of the couple was clipped to the slip gauge and the other was in close contact with the thermometer bulb. The displacement due to the gauges was calculated to the nearest  $1 \times 10^{-6}$  in ( $0.025 \mu$ ) from a knowledge of their calibration sizes at  $20^\circ \text{C}$ , the observed temperatures and the thermal expansion coefficient of the gauge material. Air temperature near the resonator was recorded from

another standard thermometer calibrated to  $0.01^\circ \text{C}$ . Barometric pressure was read to the nearest  $0.05$  mm of mercury on a Fortin instrument and vapour pressure on an Assmann psychrometer.

## RESULTS

Table 2 shows a typical set of observations and calculations for one determination of the value of  $L(\sec \theta - 1)$ . The excess fractions  $f''$  in the uppermost row are those observed in the seven krypton lines in the sequence from



to right, the piston being supported on  $G_2$  (5.5 in). Fractions obtained in the opposite sequence of reading are quoted in the next row and the mean values of  $f''$  underneath. Readings of gauge and air temperature are taken before and after the fringe observations, and the mean gauge temperature is shown in the first column. Corresponding information obtained after substitution of  $G_1$  (0.5 in) for  $G_2$  is given in the next lower section of the table. In the third section the fractions  $F$ , calculated from the nominal 5 in displacement (see table 1, column 3), are subtracted from the values of  $f$  ( $=f' - f''$ ). This operation produces the row of difference fractions ( $F$ ), underneath which is given the row of balanced orders of interference obtained by the Kösters slide rule from the difference fractions. The resulting value of the downward displacement, expressed as the error from 5 in. in micro-inches, is shown to the right.

The next three lower sections similarly yield the value of the upward displacement. It will be noted that the two values of displacement differ significantly in the two directions of movement. This is due to the differential effect of rising temperature on the expansions of the concrete pillar and the metallic resonator with its support. During rising temperatures the datum surface of the resonator, i.e. the surface of the slip gauge base, moves upwards towards the optical head. Thus the amount of downward displacement is smaller than that of an upward displacement, although the same slip gauges are interchanged. The difference in displacement at the datum surface becomes even more marked when observed at the piston error because the latter is further removed from the edge of the concrete pillar.

In the remaining sections of the table the mean displacement in the two directions is corrected for the effect of wavelength of the departures of atmospheric conditions from the standard conditions of 20°C, 760 mm and 7 mm vapour pressure, using N.P.L. data for the purpose.<sup>(5)</sup> The conditions recorded are those which

existed at the mean time of the determination, the barometric reading being reduced to the usual standard conditions of mercury at 0°C and  $g = 980.665 \text{ cm/sec}^2$ . The optically measured displacement after correction is finally compared with the mechanical displacement to derive the value of  $L(\sec \theta - 1)$ .

Table 3 shows the series of 14 individual and 7 mean values of  $L(\sec \theta - 1)$  obtained from 7 sets of observations. Set 2 is that described in more detail in Table 2. Set 3 shows a difference of only 3 micro-inches between the two directions of displacement; the temperature rise recorded during this set at one point of the resonator was only 0.05°C, and the change in mean resonator temperature was probably much smaller. In the other sets the average rise recorded at the same point was 0.15°C, individual values ranging from 0.1°C to 0.2°C, and the resulting excess of upward over downward displacement is 56 micro-inches on the average. The low mean values of sets 4 and 7 were probably due to non-uniform temperature changes in the apparatus, but they have been taken into account in deriving the final value of  $L(\sec \theta - 1)$ . It is conceivable that the distorting effect of differing temperature gradients in parts of the resonator support might cause accidental changes in the value of  $\theta$ .

The final value for  $L(\sec \theta - 1)$  of  $+12 \times 10^{-6}$  in ( $0.305 \mu$ ) is associated with a standard deviation of  $\pm 5 \times 10^{-6}$  in ( $\pm 0.125 \mu$ ) for single determinations. As the value of  $L$  was 12.7 cm (5 in) to a few parts in a million, the corresponding values of  $\sec \theta$  were  $1 + (2.4 \pm 1.0)10^{-6}$ . The values of  $\theta$  giving these values of the secant are approximately  $7\frac{1}{2} \pm 1\frac{1}{2}$  minutes of arc.

If the krypton lamp were cooled in liquid air to reduce Doppler effect in the radiations, displacements up to 30 cm could be similarly measured. Greater distances will be within the range of direct measurement when new light sources, now under development, become available; these sources employ as emitter a single isotope of even-mass number such as mercury -198 or krypton -84.

Table 3. Values of  $L(\sec \theta - 1)$

Unit =  $1 \times 10^{-6}$  in

Set	Direction of displacement	$L(\sec \theta - 1)$	
		Individual values	Mean
1	Downwards	-14	+16
	Upwards	+47	
2	Downwards	-19	+13
	Upwards	+46	
3	Upwards	+16	+15
	Downwards	+13	
4	Upwards	+31	+4
	Downwards	-23	
5	Upwards	+52	+15
	Downwards	-22	
6	Downwards	-6	+15
	Upwards	+36	
7	Upwards	+24	+5
	Downwards	-14	
Final value			$+12 \pm 5$

#### ACKNOWLEDGMENT

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# Solution of Partial Differential Equations with a Resistance Network Analogue

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[Paper first received 24 October, 1949, and in final form 15 December, 1949]

It is shown in this paper that resistance networks are capable of very high accuracy in the solution of partial differential equations with given boundary conditions, as the networks possess certain averaging properties. Fairly exhaustive tests carried out on a resistance network constructed by the author gave accuracies in the range of 1 part in 1 000 to 1 part in 10 000. The design of this resistance network is described; it is of the axially symmetrical type, with 60 meshes in the  $z$ -direction and 20 meshes in the  $r$ -direction, and is surrounded by a termination strip. Fifty current feeding points for adjusting boundary conditions are provided. The boundaries of the models need not coincide with the mesh points as correction can be made by local modifications of the network. It is also possible to take measurements within a mesh of the network and to simulate the introduction of dielectric material into an electric field. The high accuracy, and simplicity and speed of operation, make the resistance network a useful tool in the investigation of field distributions in many branches of science and engineering.

Many important problems in physics and engineering require the solution of partial differential equations of the form  $\nabla^2 f = g$ , or  $\text{div}(\epsilon \text{ grad } f) = g$ , where  $\epsilon$  is a known and  $f$  an unknown scalar function of the space co-ordinates  $x, y, z$ , for prescribed boundary values. In most practical cases, no rigorous solution can be found and a numerical solution, e.g. with the help of Liebmann's<sup>(1)</sup> iteration procedure or Southwell's<sup>(2)</sup> relaxation method, has to be obtained or experimental analogy methods such as the electrolytic tank have to be used. The numerical methods often require lengthy and tedious work if a high degree of accuracy is desired, whereas the electrolytic tank is seriously limited in its accuracy.

In the last few years, approximate experimental solutions of Laplace's and Poisson's equations were obtained by de Packh<sup>(3)</sup> and by Redshaw,<sup>(4)</sup> following proposals by Hogan,<sup>(5)</sup> with the aid of networks of electrical resistances. Estimates by de Packh of the accuracy that should be obtainable suggested that it might approach the tolerance limit of the individual resistance units, e.g.  $\pm 1\%$ . This view seemed to be confirmed by the results obtained with Redshaw's apparatus; the experimental results agreed with solutions obtained by the relaxation method using the same mesh size within a few per cent.

Thus it appeared that the accuracy obtainable with a resistance network should be of the same order as that given by a carefully used electrolytic tank. However, a fresh examination by the author of the possibilities of a resistance network, as suggested by de Packh,\* led the author to expect an altogether different order of accuracy in the solution of Laplace's equation; it also seemed to point the way to the successful combination of an iteration process with the use of current-fed resistance networks which allows one to solve a much wider range

of partial differential equations. A brief preliminary account of the new network and of the iteration method was recently given.<sup>(7)</sup>

## PRINCIPLE OF THE NETWORK OPERATION

One can look upon the resistance network analogue either as an electrolytic tank in which the continuously distributed conductance is replaced by a set of lumped conductances or as a computing machine solving certain types of difference equations for given boundary conditions. In practice it is found advantageous to employ both approaches. The first is very useful in all questions relating to the design of the apparatus and the second is more appropriate in discussions of the mathematical characteristics of the equations to be solved.

Now consider the partial differential equation

$$\text{div}(\epsilon \text{ grad } f) = g \quad (1)$$

and assume, for sake of simplicity, that we are dealing with a two-dimensional problem, depending only on  $x$  and  $y$ . Replace the differential equation by its corresponding difference equation, in which the derivatives at the point  $O$  (Fig. 1) are expressed by the differences of

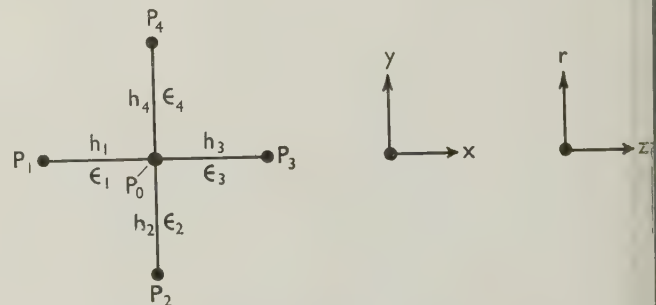


Fig. 1. Finite difference mesh points

\* The proposals of Hogan, and Redshaw's apparatus, and Kron's<sup>(6)</sup> suggestions only came to the author's notice some time after the apparatus to be described below had been completed and had been demonstrated in its operation.

the values of the function  $f$  at the neighbouring points  $P_1 \dots P_4$  and at the point  $O$ . Let the distance



$P_1$  be  $h_1$ ,  $OP_2$  be  $h_2$ , etc. The average value of the known scalar function  $\epsilon$  between  $P_1$  and  $O$  be  $\epsilon_1$ , between  $O$  and  $P_2$  be  $\epsilon_2$ , etc. Assuming that  $h_1 \dots h_4$  are so small that linear interpolation is permissible, and that the higher order terms can be disregarded, we are led to the difference expression replacing equation (1):—

$$\text{div}(\epsilon \text{ grad } f) \simeq \frac{2}{(h_1 + h_3)} \left[ \frac{\epsilon_1}{h_1} (f_1 - f_0) + \frac{\epsilon_3}{h_3} (f_3 - f_0) \right] + \frac{2}{(h_2 + h_4)} \left[ \frac{\epsilon_2}{h_2} (f_2 - f_0) + \frac{\epsilon_4}{h_4} (f_4 - f_0) \right] \quad (2)$$

If the neighbouring points  $P_1, P_2 \dots$  are all equidistant from point  $O$ ,  $h_1 = h_2 = \dots = h$ , and if, moreover, the point function is a constant,  $\epsilon_1 = \epsilon_2 = \dots = \epsilon$ , equation (2) simplifies to the familiar difference equation of the relaxation method:

$$\nabla^2 f = \frac{1}{h^2} (f_1 + f_2 + f_3 + f_4 - 4f_0) \quad (3)$$

An important class of three-dimensional fields is that of axial symmetry. In this case the field depends only on  $r$  and  $z$ , and

$$\text{div}(\epsilon \text{ grad } f) = \frac{1}{r} \frac{\partial}{\partial r} \left( r \epsilon \frac{\partial f}{\partial r} \right) + \frac{\partial}{\partial z} \left( \epsilon \frac{\partial f}{\partial z} \right)$$

this leads to the difference equation

$$\text{div}(\epsilon \text{ grad } f) \simeq \frac{2}{h_1 + h_3} \left[ \frac{\epsilon_1}{h_1} (f_1 - f_0) + \frac{\epsilon_3}{h_3} (f_3 - f_0) \right] + \frac{2}{h_2 + h_4} \left[ \left( 1 - \frac{h_2}{2r_0} \right) \frac{\epsilon_2}{h_2} (f_2 - f_0) + \left( 1 + \frac{h_4}{2r_0} \right) \frac{\epsilon_4}{h_4} (f_4 - f_0) \right] \quad (4)$$

where  $r_0$  is the distance of the point  $O$  from the axis. This simplifies for  $h_1 = h_2 = \dots = h$  and  $\epsilon_1 = \epsilon_2 = \dots = \epsilon$  to:

$$\nabla^2 f = \frac{1}{h^2} \left[ f_1 + \left( 1 - \frac{h}{2r_0} \right) f_2 + f_3 + \left( 1 + \frac{h}{2r_0} \right) f_4 - 4f_0 \right] \quad (5)$$

Now consider the application of Kirchhoff's law to the five currents  $i_1, \dots, i_4$ , and  $I$ , meeting at the junction  $P_0$  of a network of resistances (Fig. 2):

$$\sum_{i=1}^4 i_n = -I$$

but  $i_n = \frac{V_n - V_0}{R_n}$ , if the voltage applied at the point  $P_n$

is called  $V_n$ , and if the resistance in the branch  $P_n P_0$  is  $R_n$ , or

$$\frac{V_1 - V_0}{R_1} + \frac{V_2 - V_0}{R_2} + \frac{V_3 - V_0}{R_3} + \frac{V_4 - V_0}{R_4} = -I \quad (6)$$

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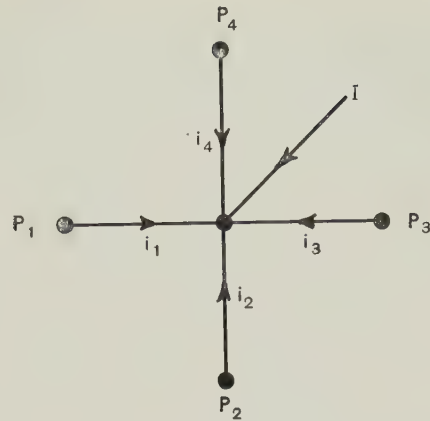


Fig. 2. Currents flowing into mesh point  $P_0$

Comparing equations (3) and (6) we see that we have a complete formal analogy between the voltages  $V$  appearing at the junctions of the resistance network and the sought function  $f_1$  if we choose the resistance values  $R_1 \dots R_4$  according to this relation for the two-dimensional case:

$$R_1 = R_2 = R_3 = R_4 = h^2 R_0 \quad (7)$$

and according to equations (8) for the axially symmetrical case:

$$\left. \begin{aligned} R_1 &= h^2 R_0 \\ R_2 &= h^2 \left( \frac{2r_0}{2r_0 - h} \right) R_0 \\ R_3 &= h^2 R_0 \\ R_4 &= h^2 \left( \frac{2r_0}{2r_0 + h} \right) R_0 \end{aligned} \right\} \quad (8)$$

and make the current  $I$  fed in at the point  $P_0$  from an external source

$$I = -g/R_0 \quad (9)$$

where  $g$  is the functional relationship given by equation (1) and  $R_0$  a suitably selected constant of the apparatus.

Thus, within the validity of the approximation of the differential equation (1) by the appropriate difference expressions, it is only necessary to set up the required boundary conditions to obtain the full solution of the equation (1) if  $g \equiv 0$  everywhere; the resistance network can therefore be considered as a machine applying the "relaxation technique" automatically and instantaneously in the case of Laplace's equation. The main effort is the extraction of the available information through the voltage measurements; this effort may be quite small, if only the knowledge of the field values at a few selected points is required, as happens in many problems.

If  $g \neq 0$ , currents as prescribed by equation (9) have to be fed in at the mesh points. In particular, if  $g$  is a

function of the sought function  $f$  itself, the iteration process mentioned in the introduction has to be applied. This will be more fully discussed in a later paper; this paper will be confined to the application of the network to the solution of Laplace's equation  $\nabla^2 f = 0$ .

#### EXPECTED ACCURACY OF THE RESISTOR NETWORK SOLUTIONS

Measurements of the distribution of the sought function  $f$  are limited in their accuracy by two fundamental factors, the mesh size, and the deviations of the network component values from their nominal design values; the voltage and current measurements required can be carried out with good enough precision to make these sources of error negligible.

*Influence of the Finite Mesh Size.* It is known from the principles underlying the relaxation method that the mesh size must be made so small that the replacement, within the specified error limit, of the original partial differential equation by the finite difference equations (3) or (5) is permissible. In practice, the number of mesh points has to be limited and it is necessary to have methods for checking the errors due to the finite mesh size.

One way of checking and of reducing these errors is the application of a method which is analogous to one given by Fox.<sup>(8)</sup> The error can be estimated by evaluating the fourth order term  $\frac{h^4}{12}(\frac{\partial^4 f}{\partial x^4} + \frac{\partial^4 f}{\partial y^4})$ , or in the axially symmetrical case

$$\frac{h^4}{12} \left[ \frac{1}{r} \frac{\partial^2}{\partial r^2} \left( r \frac{\partial^2 f}{\partial r^2} \right) + \frac{\partial^4 f}{\partial z^4} \right]$$

from the network solution, using the equations (10) or (11) (see Fig. 3):

$$\frac{h^4}{12} \left( \frac{\partial^4 f}{\partial x^4} + \frac{\partial^4 f}{\partial y^4} \right) \simeq \frac{1}{12} [(f_A + f_B + f_C + f_D) - 4(f_1 + f_2 + f_3 + f_4) + 12f_0] \quad (10)$$

and

$$\begin{aligned} & \frac{h^4}{12} \left[ \frac{1}{r} \frac{\partial^2}{\partial r^2} \left( r \frac{\partial^2 f}{\partial r^2} \right) + \frac{\partial^4 f}{\partial z^4} \right] \\ & \simeq \frac{1}{12} \left\{ \left[ f_A + \left( 1 - \frac{h}{r_0} \right) f_B + f_C + \left( 1 + \frac{h}{r_0} \right) f_D \right] \right. \\ & \quad \left. - 4 \left[ f_1 + \left( 1 - \frac{h}{2r_0} \right) f_2 + f_3 + \left( 1 + \frac{h}{2r_0} \right) f_4 \right] + 12f_0 \right\} \quad (11) \end{aligned}$$

At those mesh points where the fourth order term is not so small that it can be disregarded, the appropriate currents are then fed in at the mesh points. This is equivalent to superimposing a correction function in the relaxation technique, as was proposed by Fox.

An easier and more readily applied method of taking the fourth order term into account is the application of an extrapolation method due to Richardson.<sup>(9)</sup> If a problem is set up first on a half-scale model giving a field distribution  $f$ , and then on a full-scale model giving the distribution  $F$ , then the much more accurate distribution  $F^*$ , corresponding to a model of at least twice full scale, is obtained by equation (12):

$$F^* = F - \frac{1}{3}(f - F) \quad (12)$$

At the intermediate mesh points, which are not common to the full and the half-scale models, the values of  $F^*$  are determined from the  $F$  values by interpolation of the difference  $(f - F)$ .

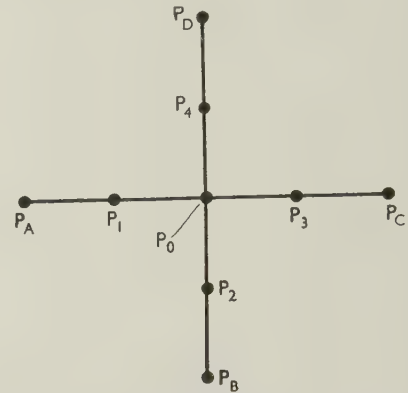


Fig. 3. Finite difference mesh points used in fourth order correction

*Influence of the Resistor Tolerances.* The second fundamental limitation in the use of a resistor network is given by the influence of the resistor tolerances on the obtainable accuracy. It will now be shown that the resistor network possesses certain averaging properties which make the results about two orders of magnitude more accurate than was predicted by the previous authors.

Let us assume that the deviations  $\delta R = \rho R$  of the actual resistance values from their nominal values  $R$  are randomly distributed and that this distribution is given by a "normal frequency curve," with a standard deviation  $\sigma = \sigma(\rho)$ . Measurements on a number of resistors used in the network described below showed that this assumption is not entirely correct, but is a good enough approximation for the purpose of this discussion.

Consider, in the two-dimensional network Fig. 4, all resistors to be of exactly the same value  $R$ ; the number of components in the  $x$ -direction be  $M$ , in the  $y$ -direction be  $N$ . If we connect all points on the left edge and all points on the right edge and apply the voltage  $V$  between them, we have a voltage drop of  $\Delta V = V_0/M$  between each successive pair of mesh points in the  $x$ -direction, whereas all points along the lines  $x = \text{constant}$  have the same potential.



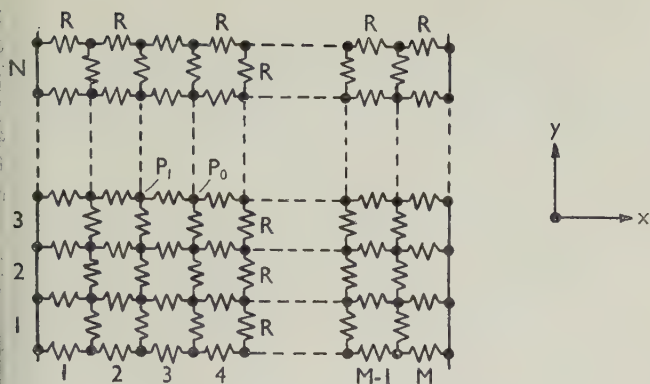


Fig. 4. Resistance network

Let us now assume that one resistor, between the points  $P_1$  and  $P_0$ , is changed by  $\delta R$  to  $R + \delta R = R(1 + \rho)$ , where  $\rho = \delta R/R$  (see Fig. 5). The voltage difference between  $P_0$  and  $P_3$  remains  $\Delta V$  as before, except for a negligibly small correction term (which may be omitted), whereas the voltage difference between  $P_0$  and  $P_1$  increases to  $\Delta V + \delta V$ , causing a change in the potential at the point  $P_0$ . In consequence, we also find new voltage differences  $\delta V'$  between  $P_0$  and  $P_2$ , and  $P_0$  and  $P_4$ . These differences will be slightly smaller than  $\delta V$ , as they cause currents to flow through the points  $P_2$  and  $P_4$  which will increase the potentials at  $P_2$  and  $P_4$  by a small amount. One finds that  $\delta V'$  lies approximately between  $\delta V$  and  $\frac{1}{2}\delta V$ , depending on the position of the point  $P_0$  within the network, being in most cases nearer to  $\delta V$ . We use therefore, as approximation, the average value  $\delta V' = \frac{3}{4}\delta V$ . The four branch currents flowing into  $P_0$  are then

$$i_1 = \frac{\Delta V + \delta V}{R + \delta R}, i_2 = \frac{\delta V'}{R}, i_3 = -\frac{\Delta V}{R}, i_4 = \frac{\delta V'}{R}$$

As 
$$\sum_{i=1}^4 i_n = 0$$

we have 
$$\frac{1}{R}[(\Delta V + \delta V)(1 - \rho) + 2\delta V' - \Delta V] = 0$$

and with fair approximation:

$$\delta V = \frac{2}{5}\rho\Delta V \quad (13)$$

or 
$$\frac{\delta V}{V_0} = \frac{2\rho}{5M} \quad (14)$$

Equations (13) and (14) express the *local* error due to the change of *one* resistance. As we had assumed the errors  $\rho$  to be statistically distributed, we can expect a "normal" error distribution for  $\delta V$ , with a standard deviation  $\sigma(\delta V)$ , obtained by replacing  $\rho$  by  $\sigma(\rho)$  and  $\delta V$  by  $\sigma(\delta V)$  in equations (13) and (14). We can easily extend our result, equation (13), to the case where both potential gradients  $\partial V/\partial x = \Delta V_x/h$  and  $\partial V/\partial y = \Delta V_y/h$  are present.

Calling the linear dimension of the model, measured in the unit  $h$ , in the  $x$ -direction (or  $z$ -direction)  $a$  and in the  $y$ -direction (or  $r$ -direction)  $b$ , we have for the average field gradients  $E_a$  and  $E_b$  in these two directions  $E_a = V_0/a$  and  $E_b = V_0/b$ . But the voltage increments between two successive mesh points in the  $x$ - and  $y$ - (or  $z$ - and  $r$ -) directions are

$$\Delta V_x = \left(\frac{\partial V}{\partial x}\right)h = E_x h \quad \text{and} \quad \Delta V_y = \left(\frac{\partial V}{\partial y}\right)h = E_y h$$

We have, therefore: 
$$\frac{\Delta V_x}{V_0} = \left(\frac{E_x}{E_a}\right)\left(\frac{h}{a}\right)$$

and 
$$\frac{\Delta V_y}{V_0} = \left(\frac{E_y}{E_b}\right)\left(\frac{h}{b}\right)$$

The more general form of equation (13) is then:

$$\sigma\left(\frac{\delta V_x}{V_0}\right) = \frac{2}{5}\left(\frac{E_x}{E_a}\right)\left(\frac{h}{a}\right)\sigma(\rho)$$

and

$$\sigma\left(\frac{\delta V_y}{V_0}\right) = \frac{2}{5}\left(\frac{E_y}{E_b}\right)\left(\frac{h}{b}\right)\sigma(\rho)$$

To obtain the *local* error of the potential at the point  $P_0$  due to the presence of tolerance errors in *all four* resistances joined in the point  $P_0$  we have to superimpose vectorially the voltage changes due to each of the four resistances. We obtain then for the standard deviation of the *local* error  $\sigma_A\left(\frac{\delta V}{V_0}\right)$ :

$$\sigma_A\left(\frac{\delta V}{V_0}\right) = \frac{2\sqrt{2}}{5}\sigma(\rho)\left[\left(\frac{E_x}{E_a}\right)^2\left(\frac{h}{a}\right)^2 + \left(\frac{E_y}{E_b}\right)^2\left(\frac{h}{b}\right)^2\right]^{\frac{1}{2}} \quad (15)$$

Next, we have to consider the effect of the statistical accumulation of the local errors in the whole network, leading to an *average* error of the field distribution which is superimposed on the local error. If we call

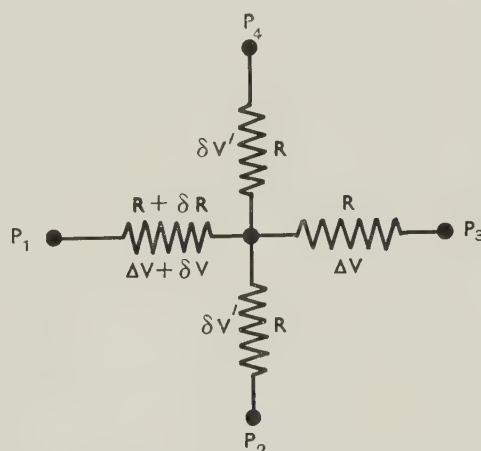


Fig. 5. Potential change due to tolerance of resistance value

$\sigma_{mn}$  the local error at the point  $P_{mn}$  where  $m = 1, 2, \dots$   
 $M = \frac{a}{h}$  and  $n = 1, 2, \dots N = \frac{b}{h}$ , each  $\sigma_{mn}$  being given  
 by equation (15), then the average error becomes

$$\sigma_B = \left( \sum_{m=1}^M \sum_{n=1}^N \frac{\sigma_{mn}^2}{MN} \right)^{\frac{1}{2}}$$

For the standard deviation  $\sigma_T(\delta V/V_0)$  of the *total error* at the point  $P_{mn}$  we have finally

$$\sigma_T\left(\frac{\delta V}{V_0}\right) = (\sigma_A^2 + \sigma_B^2)^{\frac{1}{2}}$$

This expression is cumbersome to evaluate if strong field gradients are present, but if  $\left(\frac{E_x}{E_a}\right) \simeq 1$  and  $\left(\frac{E_y}{E_a}\right) \simeq 1$ , then all the  $\sigma_{mn}$  are nearly the same and  $\sigma_B \simeq \sigma_A$ . We have therefore  $\sigma_T \simeq \sqrt{2}\sigma_A$ , bearing in mind that this underestimates the error in regions of greater than average field strength and overestimates it in regions of small relative field strength. With this proviso, we now have this final result

$$\sigma_T\left(\frac{\delta V}{V_0}\right) \simeq \frac{4}{5}\sigma(\rho) \left[ \left(\frac{E_x}{E_a}\right)^2 \left(\frac{h}{a}\right)^2 + \left(\frac{E_y}{E_b}\right)^2 \left(\frac{h}{b}\right)^2 \right]^{\frac{1}{2}} \quad (16)$$

Equations (15) and (16), whilst only approximate, are useful guides to the accuracy that may be expected in a resistance network of known tolerance limits of the components for any given type and relative size of the model under investigation. As a numerical example, let us apply our formulae to the case of an infinite parallel plate condenser,  $E_x = E_a$  and  $E_y = 0$ . For  $a = 60h$ , and  $\sigma(\rho) = 5 \times 10^{-3}$ , corresponding to the use of resistor units of  $\pm 1\%$  manufacturing tolerance, we find  $\sigma_A\left(\frac{\delta V}{V_0}\right) = 4.7 \times 10^{-5}$  and  $\sigma_T\left(\frac{\delta V}{V_0}\right) = 6.7 \times 10^{-5}$ , very small values indeed. This example represents a favourable case; for other field distributions, an accuracy in the range of 1 part in 1000 to 1 part in 10 000 can be expected for  $\sigma_T\left(\frac{\delta V}{V_0}\right)$  with this particular network, depending on the geometry and scale of the model.

It is this error-reducing statistical property of the resistance network that makes it such a useful and attractive research and design tool.

#### PROPERTIES AND DESIGN PRINCIPLES OF THE RESISTOR NETWORK

*The Plane Network.* The design procedure can be most easily derived if we consider the resistor network as the analogy of an electrolytic tank. In the two-dimensional  $x$ - $y$  tank, let the height of electrolyte be  $t$ . If we now take a small volume element of electrolyte  $t\Delta x\Delta y$  (Fig. 6), we have for the resistance of this

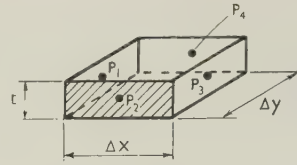


Fig. 6. Electrolytic tank analogy of resistance element

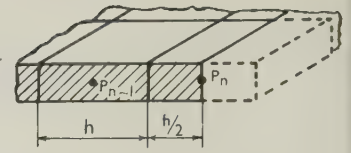


Fig. 7. Electrolytic tank analogy of resistance element at network boundary

element in the  $x$ -direction between the mesh points  $P_1$  and  $P_3$ ,

$$R_x = \frac{R_0 \Delta x}{t \Delta y} \quad (17)$$

and for its resistance in the  $y$ -direction, between the points  $P_2$  and  $P_4$

$$R_y = \frac{R_0 \Delta y}{t \Delta x} \quad (18)$$

where  $R_0$  is the specific resistance of a unit cube of electrolyte.

If the distance between points  $P_1$  and  $P_3$ :  $\Delta x = h$ , and between  $P_2$  and  $P_4$ :  $\Delta y = h$ , then  $R_x = R_y = R_0/t$ , i.e. all resistances are of equal value if we employ a mesh of squares. From Fig. 7 we can see that the resistor net-



Fig. 8. Termination surround of network

work has to be terminated by resistances of value  $2R_0/t$ , twice the standard value.

It is possible to surround the network with a "termination strip," as shown in Fig. 8, making the network effectively larger without adding too many components, and without increasing its dimensions except by a small amount. The termination resistance values can be found as follows (see Fig. 9). Let the point  $P_B$  be a

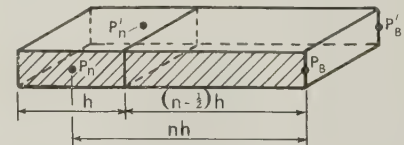


Fig. 9. Electrolytic tank analogy of termination surround

distance  $nh$  from the last point  $P_n$  within the resistor network. The resistance  $R_x$ , between  $P_n$  and  $P_B$ , is then

$$R_x = \frac{nh}{th} R_0 = n \frac{R_0}{t}, \text{ and the corresponding resistance } R_y \text{ between } P_B \text{ and } P'_B \text{ is } R_y = \frac{hR_0}{(n - \frac{1}{2})ht} = \frac{2}{(2n - 1)} \cdot \frac{R_0}{t}$$



Such a "termination strip" is very useful if problems are investigated which involve "open boundaries." A larger part of the network can then be used without danger that the reflexion of the field in the boundary of the resistor network may influence the field distribution.



Fig. 10. Electrolytic tank analogy of locally subdivided resistance network

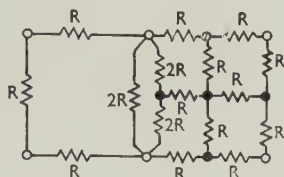


Fig. 11. Locally subdivided resistance network

It will be clear from Fig. 10 how the subdivision of the network, at places where a finer mesh is desired, can be carried out. From this we derive Fig. 11, which shows the subdivided equivalent resistor network.

It is known from the theory of electrolytic tanks that the presence of a dielectric material in an electric field, as given by the equation  $\text{div}(\epsilon \text{ grad } \phi) = 0$ , can be represented by increasing in the model experiment the depth of the electrolyte from  $t$  to  $\epsilon t$  for the space occupied by the dielectric. This principle can be applied to the resistor network; in fact, certain practical difficulties, encountered in the electrolytic tank in the application of this idea, are absent from its counterpart in the resistor network. One need only replace  $R_x$  by  $R'_x = \frac{R_x}{\epsilon}$  and  $R_y$  by  $R'_y = \frac{R_y}{\epsilon}$  for the area occupied by the dielectric material to obtain the correct field distribution.

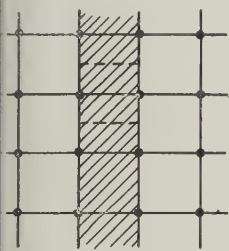


Fig. 12. Inclusion of strip of dielectric material

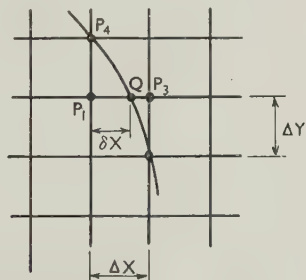


Fig. 13. Representation of curved boundaries

It is however necessary to take account of the position of the boundaries of the dielectric with respect to the resistor network junctions and to weight the modifications to the resistance values accordingly. For instance, if the dielectric occupies a strip as shown by shading in Fig. 12, then we have  $R'_x = \frac{R_x}{\epsilon}$ , but  $R'_y = \frac{2R_y}{1 + \epsilon}$ , etc. The required local changes of the network can be carried out

by temporarily shunting the network externally with appropriate resistances, without interfering with the basic network arrangement.

The method used for the derivation of the subdivided network can be applied to the representation of curved boundaries in the resistor network. If the boundaries of the model do not pass through mesh junctions only, we can modify the resistor network locally in such a way that the new mesh junctions come to lie on the (curved) boundary of the model (Fig. 13). The resistance in the  $x$ -direction is then changed from  $R_x = \frac{R_0 \Delta x}{t \Delta y}$  to  $R'_x = \frac{R_0 \delta x}{t \Delta y} = R_x \cdot \frac{\delta x}{\Delta x}$ , whereas the resistance in the  $y$ -direction will be

$$R'_y = \frac{2R_y \cdot 2R_y \frac{\Delta x}{\delta x}}{2R_y + 2R_y \frac{\Delta x}{\delta x}} = R_y \frac{2\Delta x}{\Delta x + \delta x}$$

Thus, the resistances in the  $x$ -direction are decreased in a simple relation, whereas the resistances in the  $y$ -direction are increased. The change from  $R_x$  to  $R'_x$  can be easily made by adding external shunting resistances, whereas the change from  $R_y$  to  $R'_y$  requires the replacement of the  $R_y$  resistances by those of a higher value. However, the modification of the  $R_y$ -resistances is not as important as that of the  $R_x$ -resistances, because the factor entering into the expression for  $R'_y$  is only partly determined by  $\delta x$ , and because the field gradient is, near a boundary, small in the direction parallel to the boundary. Hence, it will often lead to an acceptable accuracy in the representation of curved boundaries if only the  $R_x$ -resistances are modified, and the  $R_y$ -resistances are left unaltered.\*

Obviously, if the point  $Q$ , Fig. 13, does not lie in the  $x$ -direction from the point  $P_1$ , but in the  $y$ -direction, the required modifications of  $R_x$  and  $R_y$  become exchanged.

We can now, at least approximately, remove the only remaining drawback of the resistor network, i.e. the fact that the field distribution can only be measured at certain discrete points. For a resistor network square  $P_1 \dots P_4$  (Fig. 14) of not too large mesh size, a weighted average value for the potential at the point  $Q$ , based on linear interpolation between the potential values at  $P_1 \dots P_4$ , gives sufficiently high accuracy. This can be easily obtained by connecting a potentiometer bridge, shown diagrammatically in Fig. 15, across the points  $P_1 \dots P_4$ . The two potentiometers  $A$  and  $B$  are ganged; their tapping points are connected by the third potentiometer  $C$ . The desired potential value is measured at the tapping point of  $C$ . The potentiometers can be directly calibrated in position co-ordinates. Various tests have shown that, in this way, the potential can be measured

\* This method had already been used by Redshaw,<sup>(4)</sup> applying principles developed in the relaxation technique.

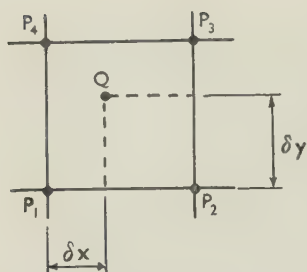


Fig. 14. Principle of potential measurement within mesh

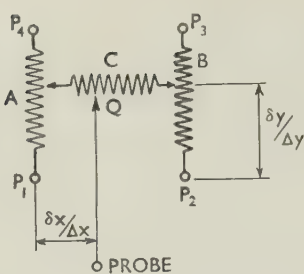


Fig. 15. Potentiometer bridge for potential measurement within mesh

within a mesh square to a position accuracy of better than one-fiftieth mesh length. This attachment is particularly useful if one wishes to determine the exact position of a given equipotential line.

*The Axially Symmetrical Network.* It is possible to construct networks for three and even four dimensions but for a given mesh size the number of components needed increases very greatly. However, three-dimensional problems of axial symmetry can be solved with a two-dimensional ( $r, z$ ) network in which the resistance values are suitably graded.

We can consider the axially symmetrical network as the analogy of an electrolytic tank with inclined bottom (Bowman-Manifold and Nicoll;<sup>(10)</sup> Pérès and Malavard<sup>(11)</sup>) where the height  $t$  of electrolyte is proportional to the distance  $r$  from the axis (Fig. 16). The resistance

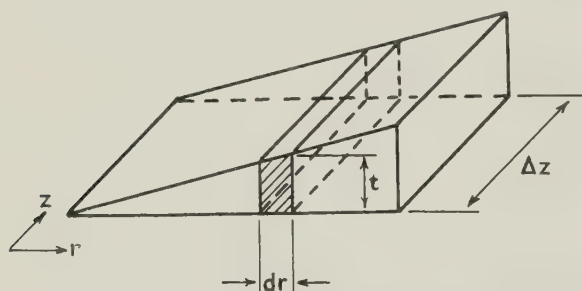


Fig. 16. Electrolytic tank analogy of resistance network for axially symmetrical problems

between two points  $P_1$  and  $P_2$ , with off-axis distances  $r_1$  and  $r_2$ , is then:

$$R(r_1, r_2) = \int_{r_1}^{r_2} \frac{R_0 dr}{t \Delta z} = \frac{R_0}{\Delta z} \int_{r_1}^{r_2} \frac{dr}{r}$$

$$\text{or} \quad R_r = \frac{R_0}{\Delta z} \log_e \left( \frac{r_2}{r_1} \right) \quad (19)$$

Similarly for  $R_z$ :

$$R_z = \frac{R_0 \Delta z}{r \Delta r} \quad (20)$$

A difficulty arises on the axis, as here  $R_z = \infty$  and  $R_r = \infty$ . This is overcome by making the line  $r = \frac{1}{2} \Delta r$  the nominal axis. Preliminary investigation of a few typical field distributions had shown that the field values at this distance from the axis have approached the truly axial values within 1 part in 10 000 for the usual size of model.

#### CONSTRUCTION OF THE RESISTOR NETWORK

The resistor network constructed by the author is of the axially-symmetrical type. The resistor components are 1 W high-stability carbon resistors of  $\pm 1\%$  manufacturing tolerance. The network provides 20 meshes in the  $r$ -direction and 60 meshes in the  $z$ -direction, requiring  $21 \times 60 + 20 \times 61 = 2\,480$  resistors. A later added termination strip extends the size of the network by 5 mesh units in the  $r$ -direction, requiring an additional 121 resistors. The resistance values are graded logarithmically, as given by equations (19) and (20), from a value of 16 000  $\Omega$  on the (nominal) axis to 100  $\Omega$  at  $r = 20$ .

The resistors are permanently assembled on a horizontal bakelite panel into which 4 B.A. screws are set in a regular array. The resistors are mounted underneath the board; the screws projecting through the bakelite panel serve as contacts for the potential measurements and, if required, as feeding points for currents injected at the mesh junctions. Models are set up on the upper side of the panel by connecting the appropriate screw studs by heavy-gauge tinned copper wire. All connexions are made by soldering to tags screwed to the studs. Numbered co-ordinate lines are engraved on the bakelite panel.

Behind the resistor network panel a vertical panel is arranged carrying 50 feeding points for injecting currents. Each feeding point comprises a voltage selector plug, two adjustable resistances for coarse and fine control and a jack socket for measuring the current fed in.

Fig. 17 shows a photograph of the apparatus; besides the resistor network itself are seen the comparison resistance bridge, a small pre-amplifier and the balanced indicating cathode-ray oscilloscope.

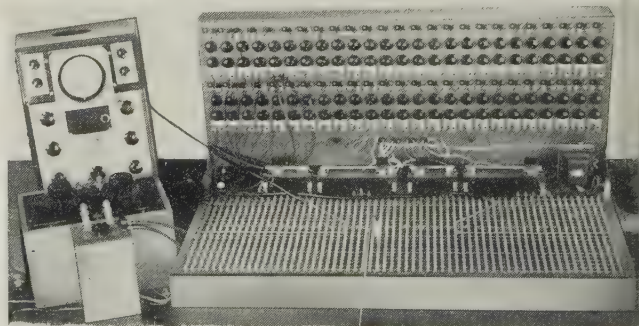


Fig. 17. Photograph of apparatus



## ELECTRICAL CIRCUITS

The circuit diagram is shown in Fig. 18. The network is fed from the 50 c/s mains, through a transformer providing  $3 \times 6$  V. The network is connected to the central 6 V section of the transformer. The two other transformer sections are used to provide negative and positive feeding voltages for current injection, and to supply two other voltages to the electrodes of the model; these are adjustable by means of two power potentiometers mounted on the bakelite panel. The high-precision potentiometer bridge is connected in parallel with the resistor network. The out-of-balance voltage is observed on a cathode-ray oscilloscope; at maximum gain of the amplifiers an out-of-balance voltage of 1 part in 100 000 can be detected.

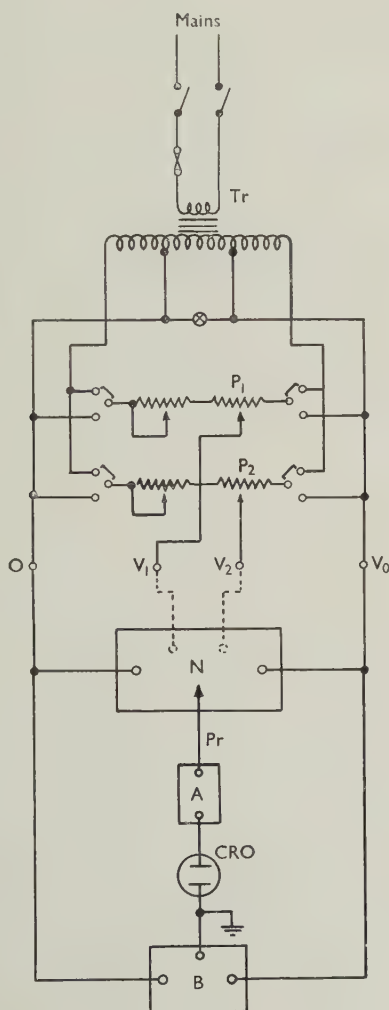


Fig. 18. Electrical circuit of apparatus

The influence of out-of-phase impedance components is negligible even at the degree of accuracy aimed at, owing to the low resistance of the network and of the measuring bridge.

## TESTS OF THE ACCURACY OF THE NETWORK

The network was tested by setting up several problems for which the solution is exactly known. The two standard problems used in routine checks, and to track down faults in the network, are the parallel plate condenser and the cylindrical condenser. Table 1 gives typical potential readings obtained for a parallel plate condenser, the plates occupying the positions  $z = 0$  and  $z = 60$ , and being at the potentials 0 and  $V_0 = 1$  respectively.

Table 1. Typical potential readings for parallel plate condenser

$r$	$z = 15$	$z = 30$	$z = 45$
0	0.25000	0.49995	0.75002
1	0.24997	0.49993	0.74999
2	0.25002	0.49988	0.74999
3	0.25000	0.49988	0.74993
4	0.24997	0.49987	0.74998
5	0.24995	0.49987	0.75000
6	0.24995	0.49990	0.75000
7	0.24997	0.49987	0.75002
8	0.24996	0.49985	0.75000
9	0.24995	0.49984	0.75008
10	0.24985	0.49985	0.75009
11	0.24993	0.49988	0.75007
12	0.24991	0.49995	0.75008
13	0.24993	0.49998	0.75006
14	0.24997	0.50000	0.75005
15	0.24999	0.50004	0.75002
16	0.24997	0.49999	0.75000
17	0.24996	0.49992	0.75001
18	0.24993	0.49994	0.75000
19	0.24990	0.49998	0.75001
20	0.24990	0.50000	0.75002
Exact value	0.25000	0.50000	0.75000

The deviations of the readings from the exact values are represented in Fig. 19, where the deviation  $\delta V$  of the average value from the exact value  $V(r, z) = z/60$  at

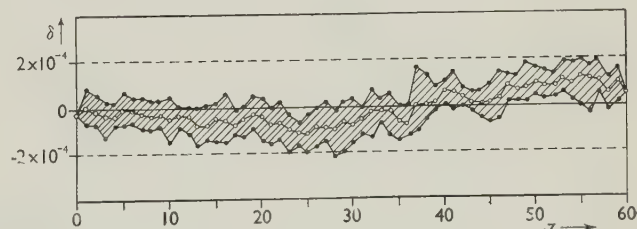


Fig. 19. Plot of extreme errors in measurement of potential distribution in parallel plate condenser

$\delta$  = deviation of measured value from exact value ( $V_0 = 1$ )  
 —○—, average value  
 —●—, maximum or minimum value } along line  $z = \text{constant}$

each position  $z = 0, 1, 2, \dots, 60$  is given by the circled central line as a function of the position  $z$ . The *extreme* deviations of the measured values from the exact values are given by the upper and lower curves in Fig. 19. It

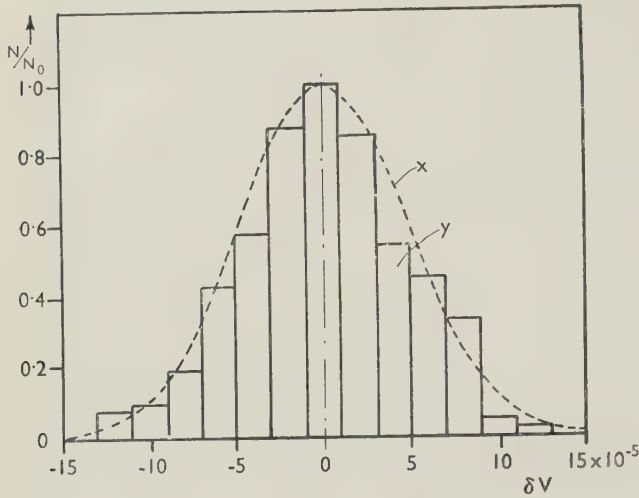


Fig. 20. Histogram showing distribution of errors in 231 measurements of potential distribution in parallel plate condenser

$$\Sigma N = 231; X, \sigma_{\text{calc.}} = 4.7 \times 10^{-5}; Y, \sigma_{\text{exp.}} = 4.5 \times 10^{-5}$$

is seen that all measured values lie with  $\pm 2.1 \times 10^{-4}$  of the exact values; only a few measured values reach these extreme deviations, and more than three-quarters of all measured points lie within  $\pm 1 \times 10^{-4}$  of the

exact values. To check the analysis leading to equations (15) and (16) in greater detail, a histogram of the distribution of the measured values was drawn (Fig. 20). This histogram represents the deviations of the individual measurements from the average value of the 21 measurements along each line  $z = \text{const.}$ , for 11 values of  $z$ , thus representing 231 measuring points. It is seen that the histogram approaches a "normal" statistical distribution curve; the "standard deviation" of the measurement  $\sigma = \sqrt{\left(\frac{\Sigma \Delta^2}{N}\right)}$  is  $\sigma_{A \text{ exp}} = 4.5 \times 10^{-5}$ , which agrees

very well with the value  $\sigma_{A \text{ calc}} = 4.7 \times 10^{-5}$ , predicted by equation (15). If the deviations are not referred to the *average* measured values, but to the exact, absolute values, then we find experimentally  $\sigma_{T \text{ exp}} = 8.5 \times 10^{-5}$ . This value represents correctly  $\sigma_{T \text{ exp}} = (\sigma_{A \text{ exp}}^2 + \sigma_{B \text{ exp}}^2)^{1/2}$ , as the independently found experimental value for  $\sigma_B$ , giving the deviation of the *average* measured values from the exact values, is  $\sigma_{B \text{ exp}} = 7.1 \times 10^{-5}$ . This value for  $\sigma_{B \text{ exp}}$  is about 50% greater than  $\sigma_{A \text{ exp}}$ , although our previous analysis would predict  $\sigma_B \simeq \sigma_A$ . This discrepancy indicates a slight systematic error in the network; it is very likely that this is due to a differential ageing effect of the carbon resistors, as one end of the network had been used preferentially for some time before these measurements were taken.

The cylindrical condenser represents a more difficult problem, as a high field strength exists near the inner conductor for a ratio of 20 : 1 of the radii of outer and inner conductors. The result of a typical test is given in Table 2; we have again very good agreement with formula 16.

Table 2. Typical potential readings for cylindrical condenser,  $r_o : r_i = 20 : 1$

$r$	Exact value	$z = 0$	$z = 10$	$z = 20$	$z = 30$	$z = 40$	$z = 50$	$z = 60$	$\sigma_T(V)_{\text{calc}}$
1	0	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	—
2	0.2314	0.2313	0.2311	0.2314	0.2319	0.2326	0.2319	0.2313	0.00067
3	0.3667	0.3673	0.3674	0.3678	0.3676	0.3677	0.3679	0.3677	0.00045
4	0.4628	0.4629	0.4629	0.4635	0.4633	0.4632	0.4634	0.4627	0.00034
5	0.5372	0.5372	0.5373	0.5372	0.5374	0.5372	0.5377	0.5375	0.00026
6	0.5981	0.5978	0.5981	0.5980	0.5981	0.5983	0.5981	0.5986	0.00023
7	0.6496	0.6492	0.6495	0.6493	0.6494	0.6496	0.6494	0.6497	0.00019
8	0.6941	0.6941	0.6940	0.6940	0.6940	0.6944	0.6942	0.6943	0.00017
9	0.7335	0.7336	0.7334	0.7336	0.7336	0.7335	0.7336	0.7337	0.00014
10	0.7686	0.7686	0.7685	0.7686	0.7686	0.7687	0.7685	0.7686	0.00013
11	0.8004	0.8003	0.8004	0.8004	0.8005	0.8005	0.8002	0.8004	0.00012
12	0.8295	0.8294	0.8295	0.8293	0.8295	0.8294	0.8294	0.8294	0.00011
13	0.8562	0.8562	0.8562	0.8562	0.8564	0.8562	0.8562	0.8560	0.00011
14	0.8810	0.8809	0.8810	0.8809	0.8811	0.8809	0.8810	0.8807	0.00009
15	0.9040	0.9040	0.9040	0.9040	0.9043	0.9040	0.9040	0.9039	0.00008
16	0.9255	0.9255	0.9256	0.9256	0.9257	0.9256	0.9255	0.9254	0.00008
17	0.9458	0.9458	0.9458	0.9458	0.9459	0.9458	0.9457	0.9458	0.00007
18	0.9648	0.9649	0.9648	0.9648	0.9649	0.9648	0.9649	0.9648	0.00007
19	0.9829	0.9829	0.9829	0.9829	0.9829	0.9829	0.9829	0.9829	0.00006
20	1	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	—



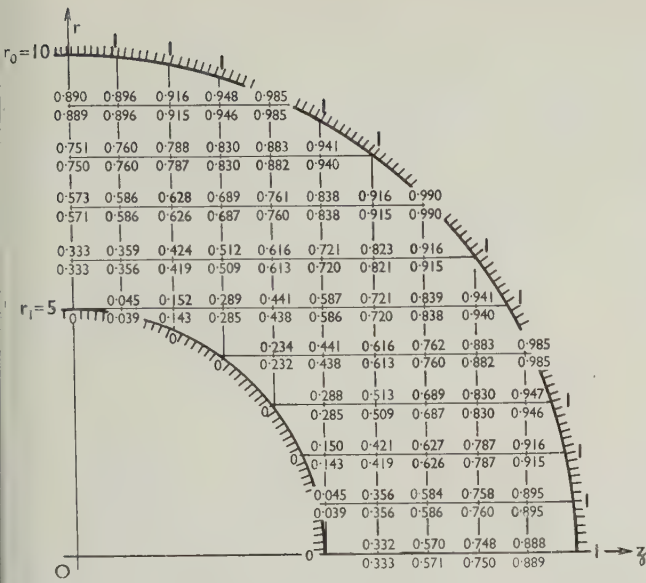


Fig. 21. Comparison of measured values and exact values of potential distribution in spherical condenser

Upper figures: Measured values. Lower figures: Exact values.

A test involving local modifications of the network by external shunts to represent curved boundaries, as described in the section on the design principles of the resistor network, is reproduced in Fig. 21. The problem is the field distribution in a spherical condenser, the inner sphere having a radius  $r_i = 5$  and the outer sphere a radius  $r_0 = 10$ . The scale of this model is quite small, involving only 5 mesh units in the radial direction. In view of this, the agreement between the measured values, standing in the upper rows, and the exact values, given in the lower rows for each mesh point, appears quite satisfactory, being within a few parts in 1 000.

To study the representation of a dielectric, a disk of height 2 and radius 10, with a dielectric constant  $\epsilon = 4$ , was placed halfway between the plates of a parallel plate condenser of plate separation 10. Table 3 gives the values of the potential along the axis, which can be calculated exactly, and the measured values. In Fig. 22 the experimentally found field distribution is plotted.

#### IMPROVEMENT OF MEASURED FIELD PLOTS THROUGH RELAXATION

As the information about the field distribution is obtained, with the resistor network, at a number of regularly arranged mesh points, it is obvious that the relaxation method can be used with advantage to check

the measured results and, if need be, to improve them. This is facilitated through the largely random distribution of errors in the network, which leads to a nearly "normal" statistical distribution curve for the residuals centred on zero residual. Hence the condition that the sum of all residuals should be small, an important convergence criterion in the relaxation technique, is automatically fulfilled in the resistor network. Moreover, experience has shown that for the measured field distributions large residuals of one sign are almost invariably surrounded by fairly large residuals of the opposite sign. It is therefore easy and quick to improve

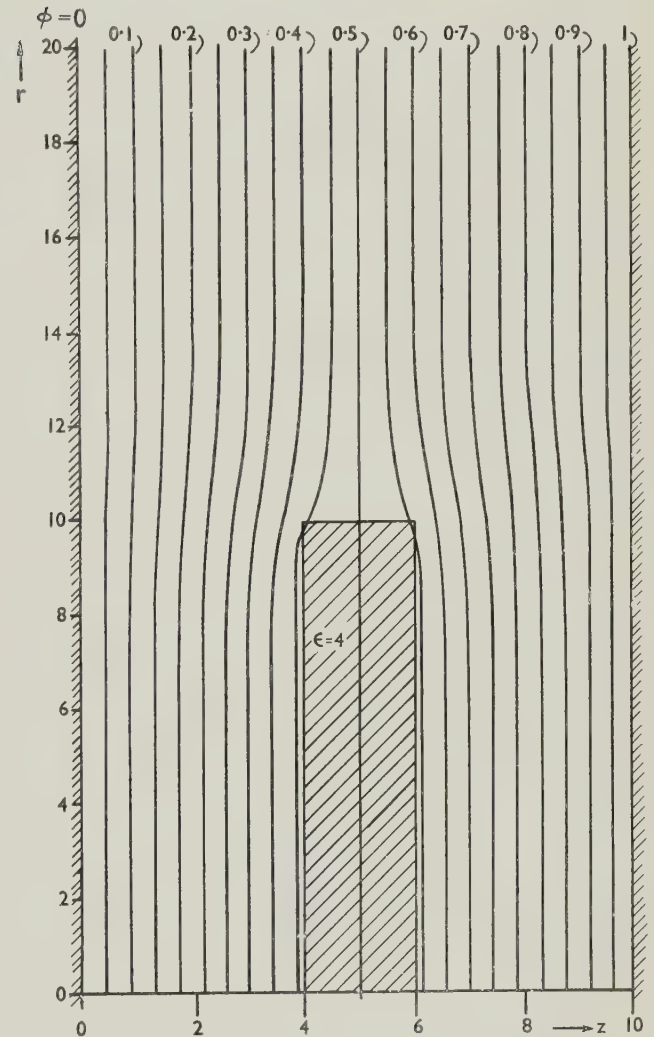


Fig. 22. Measured field distribution in parallel plate condenser containing dielectric disk

Table 3. Comparison of calculated and measured potential values along axis for parallel plate condenser containing dielectric disk

$z$	0	1	2	3	4	5	6	7	8	9	10
$\phi$ calc.	0	0.1177	0.2353	0.3529	0.4706	0.5000	0.5294	0.6471	0.7647	0.8823	1.0000
$\phi$ meas.	0	0.1179	0.2349	0.3528	0.4707	0.5002	0.5295	0.6472	0.7645	0.8820	1.0000

the accuracy of the measured field distribution through relaxation by a factor of 2 or 3, as the larger errors are localized, and the relaxation then merely consists in a smoothing out of local irregularities in the field distribution.

The effect of this improvement by relaxation in a typical case, the field distribution in an electron lens, is shown in Fig. 23 which shows the frequency distribution

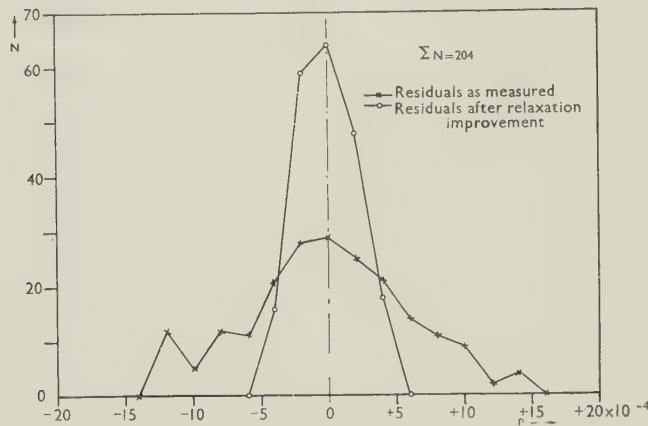


Fig. 23. Frequency diagram showing reduction of spread of residuals through relaxation (field distribution in electron lens)

of the residuals as calculated for the measured values and the frequency distribution of the residuals after relaxation had been applied to a number of points, represented by the "outer wings" of the first distribution curve.

#### TEST OF THE EFFECT OF THE FINITE MESH SIZE

To obtain an indication of the influence of the finite mesh size on the final result, and to check also the usefulness of the extrapolation formula, equation (12), the penetration of the electrostatic field through a circular aperture in an infinitely thin conducting sheet was investigated (Fig. 24). This problem was solved rigorously by Ollendorff<sup>(12)</sup> and by Fry,<sup>(13)</sup> hence a comparison of the measured and the exact solutions is possible. Moreover, the sharp edge of the aperture represents a very severe test for any numerical method employing steps of finite size. As the field in the

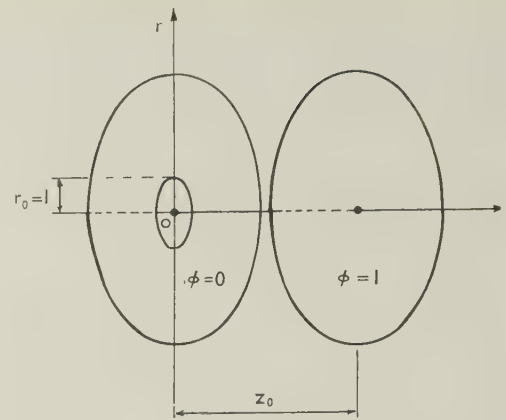


Fig. 24. Aperture in infinitely thin diaphragm

"field-free" region to the left of the aperture dies away very slowly with increasing distance from the aperture, the whole length of the resistor network was used in setting up a half-scale model. The approximate field shape at a distance of three times the aperture radius was then determined experimentally. These potential values were then used as boundary values in the setting up of the full-scale model, the boundary potentials being adjusted where necessary by feeding in current. Some typical results are given in Table 4. It is seen that the extrapolated values agree very well with the exact values except quite near the edge of the aperture. The field changes indicated by the extrapolation are by no means negligible, even near the axis.

#### CONCLUSION

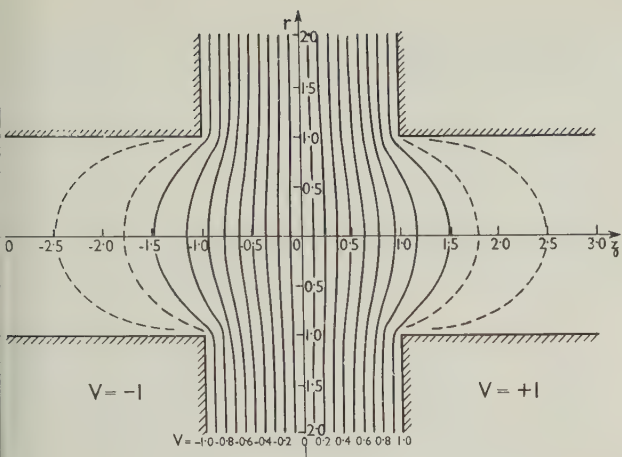
A number of field plots has been taken with the help of the resistor network described in the foregoing, and results have always been very satisfactory. A typical example is given in Fig. 25 representing the potential distribution in an electron lens. In field plotting, the resistor network proves superior to the electrolytic tank on account of its much greater accuracy and its greater simplicity of operation. Moreover, the network can be applied to more complicated mathematical problems, as will be discussed in following papers.

There seems to be little difference between the appli-

Table 4. Potentials in plane of aperture ( $z = 0$ )

$r/r_0$	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1
$\phi$ meas. { 1 : 2 scale model	0.0986	—	0.0962	—	0.0886	—	0.0744	—	0.0488	—	1
1 : 1 scale model	0.1039	0.1033	0.1017	0.0988	0.0947	0.0891	0.0819	0.0721	0.0589	0.0375	1
$-\frac{1}{3}\Delta\phi$	0.0018	(0.0018)	0.0018	(0.0019)	0.0020	(0.0022)	0.0025	(0.0029)	0.0034	(0.0044)	0
$\phi^* = \phi_{1:1} - \frac{1}{3}\Delta\phi$	0.1057	0.1051	0.1035	0.1007	0.0967	0.0913	0.0843	0.0750	0.0623	0.0419	1
$\phi$ exact	0.1057	0.1052	0.1036	0.1008	0.0969	0.0916	0.0846	0.0755	0.0634	0.0461	1
Difference	0.0000	0.0001	0.0001	0.0001	0.0002	0.0003	0.0003	0.0005	0.0009	0.0042	0





g. 25. Potential distribution in electron lens as obtained with the resistor network

ution of the resistor network and the best numerical methods for field plotting if only approximate results, ty within 1% or 2% accuracy, are required, and a very parse mesh can be used. If, however, a higher accuracy, f the order of 1 part in  $10^3$  to  $10^4$  is desired, then one btains a field plot with the resistance network in a action of the time. This is particularly so if part only f the field distribution is needed or if modifications of e shapes of models are investigated. The experimen- tal method can also be handled reliably by less killed operators.

An improved resistor network is now being designed i this laboratory using precision wire-wound resistors nd employing a test bridge of still higher accuracy than e present one. It is anticipated that, with such a network, accuracies in the range  $10^{-4}$  to  $2 \times 10^{-5}$  might e reached. Thought has also been given to methods of automatically scanning the network points and recording e field values.

The described resistor network has so far been mainly applied to electron-optical problems, but it should prove useful in many other cases where exact plots of the electric and magnetic fields in electrical machines are required and it should have applications in mechanical engineering, aerodynamics and hydrodynamics.

#### ACKNOWLEDGMENT

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# The Measurement of the Attenuation of a Longitudinal Wave Propagated along a Magnetostrictive Rod or Tube

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A new technique for measuring the attenuation of a longitudinal wave propagated along a magnetostrictive rod or tube is described. Attenuation measurements on two nickel tubes are made and the results compared with figures published elsewhere. The theory underlying the method is developed in an appendix.

A brief review of the usual attenuation measurement techniques is given in a paper by Siegel,<sup>(1)</sup> the general principle being to apply an alternating force at one end of the rod or tube and then to determine its amplitude at the other. These methods all suffer from the necessity of very careful adjustment of the generator and receiver, the possibility of direct magnetic coupling between the generator and receiver and the disadvantage of the rod or tube requiring support somewhere along the length over which the measurements take place.

The technique described in this paper utilizes the alternating magnetic field set up by a longitudinal standing wave on the tube, the magnitude of the field at any particular cross-section being proportional to the strain. Relative measurement of the field or of the voltage it induces in a small coil at three particular points along the tube then permits the attenuation to be determined. The method of measurement reduces the possibility of magnetic coupling to an absolute minimum and the influence of the support is entirely eliminated.

the measurements are to be made is located along the axis of a solenoid  $S$  by means of a support at one end. The solenoid is used to bias the tube magnetically thereby permitting measurement of the attenuation as a function of the applied magnetic field; its length should be somewhat greater than that of the tube and, in these experiments, its wound length is 126 cm. To eliminate the possibility of small alternating currents being set up in the solenoid, due to coupling with the tube, the bias current is taken through a choke of approximately 1 H. The field at the centre of the solenoid is calculated as 12 oersted/A.  $G$  is a rod or tube similar to  $T$  and is used to set up a standing wave on  $T$ ; between  $G$  and  $T$  a brass rod  $L$  is inserted, having a length of approximately 70 cm and serving to isolate the tube for measurement from the magnetic field set up by the generator exciting coil  $M$ . Excitation is obtained from a valve oscillator and amplifier. A cross-section of the connexion between the tubes  $T$  and  $G$  and the rod  $L$  is given in Fig. 2. The standing wave on the tube  $T$  gives rise to

## METHOD OF MEASUREMENT

The arrangement of the apparatus for determining the attenuation is shown in Fig. 1. The tube  $T$  on which

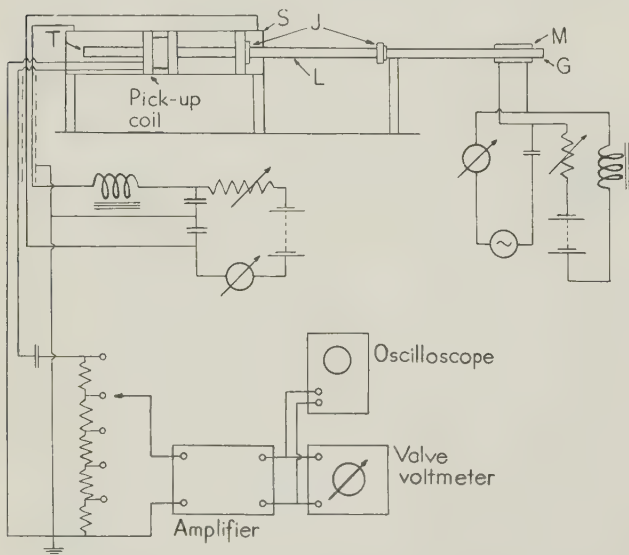


Fig. 1. Attenuation measuring apparatus

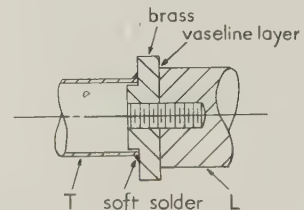


Fig. 2. Coupling between the brass rod and nickel tubes

a small alternating magnetic field whose value at any cross-section is proportional to the strain magnitude. the pick-up coil is coupled to this field, hence examination of the e.m.f. induced in the coil as it is moved along the tube permits investigation of the strain standing wave. As the e.m.f. induced in the coil is very small, it is taken to a 70 db amplifier, the output then being applied to an oscilloscope and a valve voltmeter. The pick-up coil is multi-layer wound on a wooden former; it has 280 turns and a width of 4.5 mm.

From the appendix it is seen that the attenuation may be very simply obtained from the measurement of the minimum pick-up coil voltage at two strain nodes a half wavelength apart and the maximum pick-up coil voltage at the strain antinode in between. As the incremental



magnetostriction constant is a function of the static magnetic field, it is essential that the measurements of the pick-up coil voltage are made over a portion of the tube where the flux density produced by the solenoid  $S$  is at least uniform between the two strain nodes. In addition, it is preferable to make the measurements as near as possible to the free end of the tube, as the percentage change in voltage between nodes is greatest in this region.

The actual attenuation measurement is carried out as follows. To obtain a standing wave of as great a magnitude as possible with a given excitation, the generator exciting frequency is arranged to coincide with one of the natural frequencies of the combination of tubes  $T$  and  $G$  and the brass rod  $L$ . The current to the solenoid is then set to the value under which the attenuation is to be investigated. Readings of maximum and minimum voltage, as required by the relation for determining the attenuation, are then taken over a region of the tube where the flux density is uniform.

#### EXPERIMENTAL RESULTS

Measurements were made on two soft annealed nickel tubes, each having a length of approximately 90 cm, diameter of 0.953 cm and wall thicknesses of 0.168 cm and 0.078 cm respectively. The measured values of attenuation for the two specimens are shown plotted in Fig. 3, at a frequency of 20.7 kc/s for the

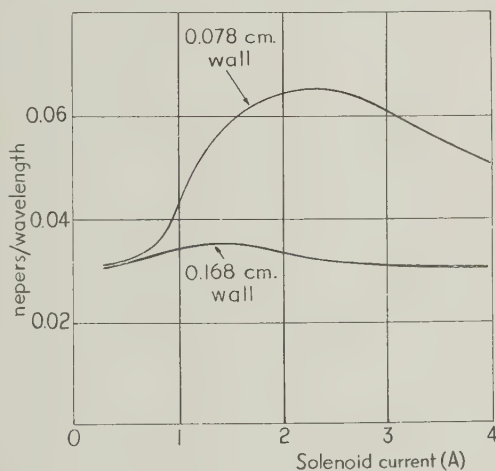


Fig. 3. Measured attenuation constant for the nickel tube specimens

tube having an 0.168 cm wall and at 21 kc/s for the 0.078 cm walled tube. The attenuation was found to be independent of the a.c. current in the exciting coil  $M$  over the range that could be employed (about 4 to 1).

Using other methods of measurement, the published values of attenuation for some nickel specimens are as follows. Wegel and Walther<sup>(2)</sup> give their result in terms of a mechanical input impedance to a half wavelength of a rod, the measurement being made with zero magnetic bias. Taking their rod diameter as 1 cm, one

finds that the attenuation is 0.0048 nepers/wavelength at a frequency of 21 kc/s. Siegel and Quimby,<sup>(3)</sup> making measurements at 36 kc/s on a rod 0.462 cm in diameter, found that the attenuation varied from 0.05 to 0.001 nepers/wavelength as the magnetic bias was taken from zero to saturation. Ide<sup>(4)</sup> gives results for magnetostrictive materials which, in some cases, show increasing loss as the magnetic bias is increased whilst in other cases the reverse is found. Although no direct comparison can be made between the results of these experiments and the above published values, it would seem that the figures obtained are of the correct order.

With regard to the variation of the attenuation with the magnetic bias, it would be expected that this is chiefly due to the change in the loss due to eddy currents set up by the small alternating field. A theoretical analysis of the attenuation due to eddy currents shows that at any particular frequency the loss is proportional to  $\lambda^2/\mu^{3/2}$ , where  $\lambda$  and  $\mu$  are respectively the incremental magnetostriction constant and the incremental permeability for the tubes.

#### CONCLUSION

A new method of measuring the attenuation of a longitudinal wave propagated along a magnetostrictive rod or tube has been devised and results obtained for two nickel tubes. The method of measurement is somewhat simpler than the usual techniques and the necessity of supporting the rod or tube along the length under measurement is avoided; magnetic bias is easily applied, but it is essential that the field is uniform over the region on which the measurements are made. Coupling between the pick-up coil and the generator is eliminated by the insertion of a brass rod between the generator and receiving tubes. The measurements were carried out as part of some general experiments on magnetostriction ultrasonic generators.

#### ACKNOWLEDGMENT

The writer desires to express his gratitude to Professor Carter for assistance in the work and to Imperial Chemical Industries Ltd. for their Fellowship.

#### APPENDIX

##### *The Theoretical Basis of the Attenuation Measurements*

The method of measurement is based on the alternating magnetic field set up by a longitudinal standing wave on the magnetostriction rod or tube. The pick-up coil and the rod or tube on which the measurements are to be made are shown in Fig. 4, one end of the tube being

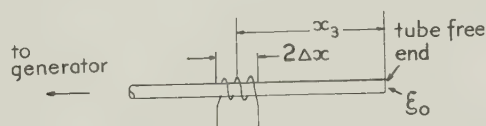


Fig. 4. Tube for attenuation measurements

free and the other coupled to a longitudinal wave generator. As the tube end is free and is therefore, in effect, terminated by zero mechanical impedance, the velocity  $\dot{\xi}$  at any cross-section distance  $x$  from the free end will be given by

$$\dot{\xi} = \dot{\xi}_0 \cosh(\alpha + j\beta)x \quad (1)$$

where  $\dot{\xi}_0$  is the velocity at the free end of the tube and  $\alpha$  and  $\beta$  are the attenuation and phase constants respectively. Due to the reverse magnetostriction, or Villari effect,<sup>(5)</sup> it follows that the voltage induced in the pick-up coil as a result of the strain  $(\partial\xi/\partial x)$  may be written

$$E = D \int_{x_3 - \Delta x}^{x_3 + \Delta x} \frac{d(\frac{\partial \xi}{\partial x})}{dt} dx \quad (2)$$

where  $D$  is a constant,  $x_3$  is the distance from the tube free end to the centre of the pick-up coil and  $\Delta x$  is half the coil length.

Carrying out the integration of equation (2),

$$E = D \left( \dot{\xi} \Big|_{x=x_3+\Delta x} - \dot{\xi} \Big|_{x=x_3-\Delta x} \right) \quad (3)$$

writing,  $f(x) + jg(x) = \cosh(\alpha + j\beta)x$

the components of the pick-up coil voltage may be obtained as follows

$$e_1 = D[f(x_3 + \Delta x) - f(x_3 - \Delta x)] \quad (4)$$

$$e_2 = jD[g(x_3 + \Delta x) - g(x_3 - \Delta x)] \quad (5)$$

For  $\beta\Delta x$  very small, then equations (4) and (5) become

$$e_1 = 2\Delta x D f'(x_3)$$

$$e_2 = 2j\Delta x D g'(x_3)$$

Substituting for  $f'(x_3)$  and  $g'(x_3)$ , one finds that

$$e_1 = 2\Delta x D(\alpha \sinh \alpha x_3 \cos \beta x_3 - \beta \cosh \alpha x_3 \sin \beta x_3)$$

$$e_2 = 2j\Delta x D(\beta \sinh \alpha x_3 \cos \beta x_3 + \alpha \cosh \alpha x_3 \sin \beta x_3)$$

Assuming that the attenuation is small, so that

$$\cosh \alpha x_3 = 1$$

$$\sinh \alpha x_3 = \alpha x_3$$

the above equations become

$$e_1 = 2\Delta x D(\alpha^2 x_3 \cos \beta x_3 - \beta \sin \beta x_3) \quad (6)$$

$$e_2 = 2j\Delta x D(\beta \alpha x_3 \cos \beta x_3 + \alpha \sin \beta x_3) \quad (7)$$

In the absence of loss ( $\alpha = 0$ ) it is obvious from equations (6) and (7) that the output voltage from the pick-up coil is zero when  $\beta x_3 = n\pi$  where  $n$  is an integer. The minimum value of pick-up coil voltage will now be determined when loss is present; clearly this will occur

in the vicinity of  $x_3 = n\pi/\beta$ . Suppose  $\beta x_3 = n\pi + \theta'$  where  $\theta'$  is very small, hence

$$\sin \beta x_3 = (-1)^n \theta'$$

$$\cos \beta x_3 = (-1)^n$$

Substituting in equations (6) and (7)

$$e_1 = 2\Delta x D(-1)^n \left( \alpha^2 \frac{n\pi}{\beta} - \beta \theta' \right) \quad (8)$$

$$e_2 = 2j\Delta x D(-1)^n (\alpha n\pi) \quad (9)$$

Hence the minimum voltage from the pick-up coil is

$$E_{min} = 2j\Delta x D(-1)^n \alpha n\pi \quad (10)$$

The maximum voltage from the pick-up coil will now be investigated; with no loss present the voltage would be a maximum when  $\beta x_3 = (2n+1)\pi/2$ . Suppose  $\beta x_3 = (2n+1)\pi/2 + \theta''$  where  $\theta''$  is very small and therefore the following approximations may be made

$$\sin \beta x_3 = (-1)^n$$

$$\cos \beta x_3 = (-1)^{n+1} \theta''$$

Substituting in equations (6) and (7),

$$e_1 = 2\Delta x D(-1)^n \left[ -\alpha^2 \left( \frac{2n+1}{2} \right) \frac{\pi}{\beta} \theta'' - \beta \right] \quad (11)$$

$$e_2 = 2j\Delta x D(-1)^n \left[ -\alpha \left( \frac{2n+1}{2} \right) \pi \theta'' + \alpha \right] \quad (12)$$

Hence the maximum voltage from the pick-up coil is

$$E_{max} = 2\Delta x D(-1)^n (-\beta + j\alpha)$$

As usually  $\beta \gg \alpha$  the above may be written

$$E_{max} = 2\Delta x D(-1)^{n+1} \beta \quad (13)$$

Therefore, from equations (10) and (13),

$$\frac{\left| \frac{E_{min}}{x_3 = (n+1)\pi/\beta} \right| - \left| \frac{E_{min}}{x_3 = n\pi/\beta} \right|}{|E_{max}|} = \frac{\alpha\pi}{\beta} = \frac{\alpha\Lambda}{2} \quad (14)$$

where  $\Lambda$  is the wavelength in the tube.

Thus, by taking measurements of maximum and minimum output voltage over a portion of the tube, the attenuation per wavelength may be found.

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# Note on the Rheological Properties of Elasto-Plastic Materials

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It is shown that the flow properties of some elasto-plastic materials can be expressed by an empirical equation. Three constants, derived from this equation, are a measure of these properties.

Bingham<sup>(1)</sup> has shown that, for many materials, the state of deformation under an applied stress can be divided into two zones, a zone of Hookean elasticity and a zone of linear flow, i.e. a zone which shows a linear relation between applied stress and rate of deformation, the transition point between the zones being called the yield point of the material. Such materials are also described as Bingham bodies or elastoplastic materials. Many investigators have found that, for a number of materials, there is no sudden transition point, but an intermediate zone in which the deformation follows neither elastic nor linear flow conditions. Bingham bodies usually show a small zone of non-linear flow if the critical shear stress has been exceeded in only a part of the field of flow (e.g. Reiner<sup>(2)</sup>). In some materials, however, which may be termed generalized Bingham bodies (Reiner<sup>(3)</sup>), the intermediate zone extends to high values of shear stress (e.g. Houwink<sup>(4)</sup>) and rheological constants are required to describe the flow properties of these materials.

## EXPERIMENTS

Goldsmith and Iball<sup>(5)</sup> have described a torsion device for the measurement of the rheological properties of visco-plastic materials which, by timing the displacement of a small vane attached to a torsion wire, permits the plotting of displacement ( $\delta$ ) — time ( $t$ ) curves for a varying applied shear stress  $\tau$ . The Bingham curve of the rate of shear ( $D = d\sigma/dt$ ,  $\sigma$  denoting the shear) or of the rate of rise of the  $\delta - t$  curve ( $d\delta \propto d\sigma$ ), against  $\tau$  can thus be plotted from a single torsion test. Such  $D - \tau$  diagrams for two typical gels formed by 1% and 1.5% solutions of commercial soaps and for a chalk and gum paste are given in Fig. 1.

In this torsion device, the initial stress at zero time is a maximum and the stress decreases with increasing time. Stress and shear strain are interdependent, as both are a function of the displacement  $\delta$  and, with only two variables,  $\delta$  and  $t$ , three-dimensional representation of stress, shear strain and time is not necessary. The  $D - \tau$  curves show that, initially, the deformation of the gels and the paste appears to follow linear flow, but it changes rapidly to non-linear flow with decreasing stress.  $D$  reaches the  $\tau$ -axis at the true yield stress  $\tau_F$  when flow ceases and  $t$  approaches infinity. The  $D - \tau$  diagrams thus describe the behaviour of visco-plastic

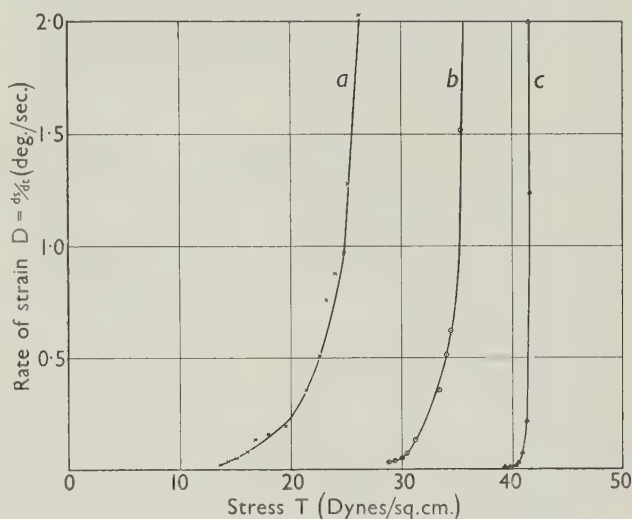


Fig. 1. Relation between rate of strain and stress (a) chalk and gum paste; (b) 1% soap gel; (c) 1.5% soap gel

materials with  $\tau_F > 0$ . These materials are, therefore, elasto-plastic.

## DISCUSSION

Scott Blair and Coppen<sup>(6)</sup> established the validity of an equation proposed by Nutting,<sup>(7)</sup> in which the shear is related to a power  $\beta$  of a constant stress  $S$  by,

$$\sigma = S^\beta t^k / \Psi \quad (1)$$

They defined the constants  $k$  and  $\Psi$  as the dissipation coefficient and the firmness intensity factor respectively. Scott Blair later showed that this equation also holds in many cases for relaxation at constant strain.

When differentiated with respect to time, equation (1) becomes,

$$D = d\sigma/dt = \frac{1}{\Psi} (\beta S^{\beta-1} t^k dS/dt + k t^{k-1} S^\beta) \quad (2)$$

This is the general differentiated form of the Nutting-Scott Blair equation when the stress varies with time but it is not valid when  $\sigma$ ,  $S$  and  $t$  are varying together. If the stress is constant, equation (2) can be written,

$$D = k t^{k-1} S^\beta / \Psi \quad (3)$$

Goldsmith and Iball have found that the rate of shear of visco-plastic substances, such as milk curd, follows an

equation of similar form to equation (3). As, in their experiments, the stress varied with time, different constants should be used to avoid confusion between their equation and the Nutting-Scott Blair equation. Hence,

$$D = at^{a-1}S_{(t)}^b/\zeta \quad (4)$$

where  $a$ ,  $b$  and  $\zeta$  are constants describing certain rheological characteristics of the material, and  $S_{(t)}$  is the stress which is dependent on time. Scott Blair<sup>(8)</sup> has pointed out that such characteristics are not physical properties of the materials in the usually accepted sense of the term. The constant  $\zeta$  has dimensions which depend on the magnitudes of the constants  $a$  and  $b$ , so that samples can be compared only in terms of the three constants,  $a$ ,  $b$  and  $\zeta$ . Equation (4) is thus a new equation, empirically derived from the observed deformation of visco-plastic substances under a varying applied stress, where the variations of both stress and shear with time are interdependent. The equation describes the flow conditions of these substances,  $S_{(t)}$  being the "flow" stress. As the minimum value of  $S_{(t)}$  occurs when  $\tau = \tau_F$ , i.e. when flow ceases,

$$S_{(t)} = \tau - \tau_F \quad (5)$$

It has been shown that, with the torsion head turned through an angle  $\alpha$ , at any displacement  $\delta$ ,  $\tau = C_1(\alpha - \delta)$ , and hence  $\tau_F = C_1(\alpha - \gamma)$ ,  $\gamma$  being the displacement at infinite time and thus at the yield point, and  $C_1$  an instrument constant. Thus,

$$S_{(t)} = C_1(\gamma - \delta) \quad (6)$$

and equation (4) becomes

$$D = C_1^b(\gamma - \delta)^b at^{a-1}/\zeta \quad (7)$$

This equation is now suggested to describe the relation between  $D$  and  $\tau$  for an elasto-plastic material, i.e. a generalized Bingham body, under the experimental conditions outlined above.

The integrated form of equation (7) can be written,

$$\gamma^{(1-b)} - (\gamma - \delta)^{(1-b)} = (1-b)C_1^b t^a / C_2 \zeta \quad (8)$$

assuming that  $d\sigma = C_2 d\delta$ , or, if  $b = 1$ ,

$$\ln[\gamma/(\gamma - \delta)] = C_1 t^a / C_2 \zeta \quad (9)$$

The logarithmic form of equation (9) is plotted in Fig. 2 in terms of the relation between  $\log \ln[\gamma/(\gamma - \delta)]$  and  $\log t$ . This relation is seen to be linear and equation (9) is thus shown to be valid for the gels and pastes tested where  $D$  and  $\tau$  have a common variable, the displacement  $\delta$ , so long as the index of stress is unity.

The factor  $a$  can be evaluated graphically from the slope of the  $\log \ln[\gamma/(\gamma - \delta)] - \log t$  line, and  $\zeta$  from its intercept with the  $\log \ln[\gamma/(\gamma - \delta)]$  axis. This intercept is equal to  $C_1/C_2 \zeta$ .  $C_1$  and  $C_2$  are instrument constants. The ratio  $C_1/C_2$  can be determined experi-

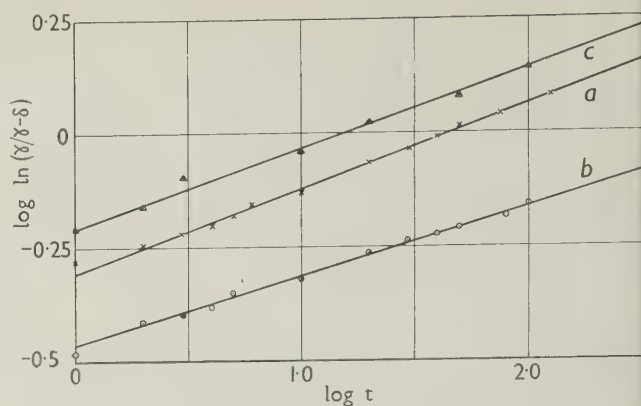


Fig. 2. Relation between  $\log \ln(\gamma/(\gamma - \delta))$  and  $\log t$  (a) chalk and gum paste; (b) 1% soap gel; (c) 1.5% soap gel

mentally by using a substance of known viscosity  $\eta$ , in which case  $\eta = \zeta$ , and  $a = 1$ .

Herschell and Bulkley<sup>(9)</sup> have proposed an equation for quasi-plastic flow which may be written,

$$D = (\tau - \tau_F)^b / \eta^* \quad (10)$$

The flow coefficient  $\eta^*$  can be obtained in terms of the constant  $\gamma$  from equations (7) and (10), so that

$$\eta^* = \zeta t^{1-a} / a \quad (11)$$

If  $b = 1$ , equation (10) becomes identical, in the main, with Bingham's equation, and

$$D = (\tau - \tau_F) / \eta_{PL} \quad (12)$$

where  $\eta_{PL}$  is the "plastic viscosity," or the inverse of Bingham's "mobility," and where the Bingham yield value  $\tau_B$ , given by the intercept of the extrapolated linear part of the  $D - \tau$  curve with the  $\tau -$  axis, is replaced by the true yield value  $\tau_F$ . Similar equations, such as that established by Reiner and Rivlin<sup>(10)</sup> for the behaviour of a Bingham body in a rotation viscometer, can be expressed in the same terms as equation (12). Again, from equation (12), and equation (7) for  $b = 1$ ,

$$\eta_{PL} = \zeta t^{1-a} / a \quad (13)$$

The relation between the flow coefficients  $\eta^*$  and  $\eta_{PL}$  and the constant  $\zeta$  is thus independent of the index of stress, but it is a function of time.  $\eta^*$ , or  $\eta_{PL}$ , and  $\zeta$  are equal only for materials with linear flow characteristics, where  $a = 1$ . For non-linear flow characteristics, where the intermediate zone is large, and where  $a < 1$ , the flow coefficients are determined by the tangents to the  $D - \tau$  curves at any given point. As the flow coefficients must, therefore, vary from point to point, they cannot be used to describe the rheological properties of those elasto-plastic (generalized Bingham) materials for which most of the deformation extends over the intermediate zone.

Many such materials have been reported in the



literature. Apart from gels (Fig. 1), they include stiff pastes (Ward and Freeman<sup>(11)</sup>), solid-in-liquid dispersions (Williamson<sup>(12)</sup>) and polymerized substances (Houwink<sup>(13)</sup>). The mechanism of the deformation of gels at low values of shear stress has been explained by Houwink<sup>(13)</sup> in terms of an increasing disruption of the micelle structure (rheodestruction) and, with complete disruption at high values of shear stress, an approach to linear flow is to be expected. The deformation of pastes can similarly be attributed to an increasing disruption of the binding forces between individual particles. This is confirmed by the shape of the  $D - \tau$  diagrams. For large values of  $\tau$ , the description of the flow properties in terms of flow coefficients may be adequate, but a means of assessing the rheological properties at low values of  $\tau$  is required. The constants  $a$ ,  $b$  and  $\zeta$  are, therefore, put forward to describe rheological characteristics of elasto-plastic materials for any form of their flow characteristic under conditions of shear.  $a$ ,  $b$  and  $\zeta$  are necessary to characterize the flow properties of a material under a decreasing stress which is dependent on the shear, but  $b$  was found, however, to be unity for a number of gels and pastes.

Some practical difficulty is experienced in evaluating the deformation at the yield point,  $\gamma$ , as the characteristics of elasto-plastic materials often change with ageing, and as the yield point is approached only very gradually, so that the deflexion  $\gamma$  at  $t \rightarrow \infty$  cannot be observed. Goldsmith<sup>(14)</sup> has explained that, by the torsion method, elasto-plastic materials can be subjected to instantaneous elastic deformation, the maximum of which gives the yield deflexion  $\gamma$ .

#### CONCLUSIONS

It has been shown that the flow properties of some elasto-plastic (generalized Bingham) materials, such as certain gels and pastes, can be expressed by an empirical equation if the materials are subjected to a varying stress which is dependent on the shear. Three constants, which can be derived from this equation, serve to characterize the flow properties. The work discussed has been limited to soap gels and chalk pastes, but the results may apply to other materials which exhibit

appreciable non-linear flow characteristics. The constant  $\zeta$  has been found of value in determining the ageing of certain gels and in comparing the flow properties of certain pastes, where neither  $a$  nor  $b$  varied substantially from case to case. In general, however, materials can only be compared in terms of the three constants  $a$ ,  $b$  and  $\zeta$ .

#### ACKNOWLEDGMENT

The author is indebted to Dr. G. W. Scott Blair for his helpful advice and suggestions, to Dr. J. Iball for his comments on this paper and to Miss B. A. Edkins for her help in the experimental work.

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## NOTES AND NEWS

## New Books

**Colours and How We See Them.** By H. HARTRIDGE, M.A., M.D., Sc.D., M.R.C.P., F.R.S. (London: G. Bell and Sons, Ltd.) Pp. xi + 158. Price 15s. net.

The Royal Institution Christmas Lectures of 1946-47, given by Professor Hartridge, have provided the material for this book. Three chapters on vision, colour illusions, and what may be called colour conjuring, are rich in information and entertainment value; the remaining three chapters condense into a small space a large number of more conventional colour experiments. It is evident that the lectures were fine and the choice of material inspired; but the book itself is too compressed to explain all the demonstrations that it recreates so vividly. Thus, on plate 2J it appears as if saturation and hue have been confused; very careful study, unaided by the relevant four lines of text, eventually shows the picture to be correct. Three-colour printing is discussed in eight lines, with reference to a rather poor plate in which the Red Queen appears very browned off. There are, indeed, some topics, such as the aurora and colour television, which are listed on the jacket but, if mentioned in the text at all, figure so briefly that they elude a fairly deliberate search for them.

There are several errors which, though perhaps trivial to the general reader, deserve notice here. Plate 3A gives a spectrum of a draining soap film which is geometrically improbable and disagrees with the colours of the film beside it. The phrases "low-colour temperature" and "higher-colour temperature" read strangely. "Accommodation" is used in one place for "adaptation." Nickel prisms exist, and were used in one famous experiment—but not for producing polarized light!

Whilst judging the book as a whole to be a little below the standard of the best "classics of popular science," the reviewer thoroughly enjoyed it and can recommend it both for the wealth of material it contains and for the happy style in which the author reproduces the atmosphere of the original lectures.

G. R. NOAKES

**Recent Advances in Radio Receivers.** By L. A. MOXON. (London: Cambridge University Press.) Pp. ix + 183. Price 18s. net.

This is one of a series of monographs dealing with advances in radio technique, and can be thoroughly recommended to those interested in the science of radio who want an up-to-date survey of the latest progress in the field of reception. It well bears out the publishers' claim for the series that the theoretical treatment is essentially physical, elaborate mathematics being excluded. The main emphasis of the book is on those factors which affect the operation of receivers under weak signal conditions, the problem of receiver "noise" receiving particular attention; and it is therefore hardly at all concerned with normal broadcast receivers, but mainly with those for radar and communication purposes.

D. H. BLACK

**A Symposium on Electronics.** Edited by A. G. PEACOCK, B.Sc., A.R.I.C., A.Inst.P. (London: Chapman and Hall, Ltd.) Pp. xiii + 199. Price 16s. net.

This well-produced book, of 200 pages, is published in collaboration with the Scientific Instrument Manufacturers' Association of Great Britain and forms a valuable record of ten papers read at the first symposium on Electronics in Scientific Research and Industry, held in London in November 1948. The papers are broadly divided into two sections as follows:

*Electronics in Research:* Electronics in Computing—D. J. Mynall; Frequency Measurements—E. A. R. Palmer and R. W. Darvill; The Measurement of Small Displacements by Electrical and Electronic Methods—R. E. Reason; The Measurement of Ionizing Radiation—G. Syke; High Vacuum Gauges—N. Steckelmacher; The Radiosonde—J. F. Veevers.

*Electronics in Industry:* Some Industrial Applications of Ultrasonics—D. O. Sproule; Metal Detection in Industry—G. S. Elphick, A. R. Woods, S. Y. Logan; Electronics in Spectroscopy—F. Holmes; Some Developments in Picture Telegraphy—J. Bell.

The publication of these papers will be greatly welcomed by all who are interested in the rapidly expanding field of electronics and its industrial applications.

S. RODDA

**On the Systems Formed by Points Regularly Distributed on a Plane or in Space.** By M. A. BRAVAIS. (Irrington-on-Hudson: The Crystallographic Society of America.) Pp. viii + 113. Price \$3.90.

Although this classical work on crystallography was originally published one hundred years ago, this volume, the first of a series of memoirs to be published by the Crystallographic Society of America, is the first English translation. It contains a number of theorems on the geometrical properties of rows and planes defined by a lattice of points in space considered in relation to the several symmetry classes which may be distinguished and the argument leads up to the deduction of the fourteen distinct types of space lattice. It is of interest also to note that, in this memoir, Bravais put forward the idea of a polar lattice which may be considered to be the forerunner of the reciprocal lattice which is of such importance in modern X-ray crystallography.

A. E. DE BARR

**Aerials for Metre and Decimetre Wavelengths.** By R. A. SMITH, M.A., Ph.D., A.M.I.E.E. (London: Cambridge University Press.) Pp. xi + 218. Price 18s. net.

The foundation of aerial design is well covered in this book. The theory of the half-wave dipole is discussed at some length and proofs are given of the equivalence of polar diagrams and self-impedance of aerials for transmission and reception in free space. The author carefully points out that the current distributions in the two cases are not in general the same and that the equivalence relations do not hold in a



on-linear propagation medium, points which are seldom appreciated. Chapter 15, on noise in aerials, though short, is very important, since it indicates how efficiency requirements and matching conditions may differ for the transmitting and receiving cases.

On the practical side, many approximate mathematical formulae, tables and diagrams of aerial performance data are given, much of which has previously been inaccessible to the general reader, together with many practical hints contained in descriptions of some aerial designs for metric wavelengths selected to be representative of principles rather than of applications.

The book appears to contain very few errors, though perhaps the array described on p. 15 as "biconical" is better known as "binomial."

W. A. JOHNSON

#### **Tables of Scattering Functions for Spherical Particles.**

(Washington: U.S. Government Printing Office.)

Pp. 119. Price 60c. post paid.

Based on Mie's theory on the scattering of light by particles with a radius comparable in magnitude with the wavelength of light. Gives the angular distribution of intensity and the total light scattered by a small spherical particle in terms of size of particle and wavelength of incident light.

#### **Handbook of Industrial Radiology (2nd edition).**

Edited by PROFESSOR J. A. CROWTHER, M.A., Sc.D.,

F.Inst.P. (London: Edward Arnold and Co.)

Pp. 218. Price 21s. net.

This handbook had its origin in a series of lectures given before the Industrial Radiology Group of The Institute of Physics. The nine chapters, each contributed by a specialist, are: The Physical Principles of Industrial Radiology; Requirements in Design of Industrial X-ray Equipment; Quantitative Measurements in Industrial Radiology; The Response of Photographic Materials to X-rays; The Radiography of Heavy Metals; The Radiography of Light Metals;

Gamma Radiography; Some Uncommon Applications of Industrial Radiography; X-Ray Protection.

In this second edition the information has been brought up to date, the most conspicuous changes being in the chapters on industrial equipment and on photographic materials. An appendix giving linear absorption coefficients has been added.

Once again the editor and authors are generously passing the royalties on the sale of the book to The Institute of Physics Benevolent Fund.

#### **Photograms of the Year, 1950.**

(London: Iliffe and

Sons, Ltd.) Pp. 20 and 68 plates. Price 7s. 6d.

net, paper cover, 10s. 6d. net, cloth binding.

This, the fifty-fifth edition, presents a selection of the world's photographic art. The plates, containing 79 reproductions of outstanding recently exhibited pictures, cover a wide variety of artistic subjects from many countries. The introduction is by Mr. C. Wormald, President of the Institute of British Photographers, and the notes on the year's work, with commentaries on the pictures reproduced in the volume, are by Mr. B. Sinkinson.

#### **F.B.I. Register of British Manufacturers—1949–50.**

22nd edition. (London: Kelly's Directories Ltd.,

and Iliffe and Sons, Ltd.) Pp. 807. Price 42s. net.

The 1949–50 Register gives details of the Organization and lists about 6000 firms and their products. In the Products and Services section manufacturers are classified under industry headings which assists rapid identification of sources of supply. The Addresses section gives complete information on every member. The Brand and Trade names and Trade marks section helps the user to trace the makers and products when only the brand or trade name is known. The Register is an authoritative guide to sources of supply and continues to be of great value to customers and manufacturers at home and overseas.

## **Notes and Comments**

### **Elections to The Institute of Physics**

The following elections have been made by the Board of the Institute of Physics:

**Fellows:** E. C. Bullard, W. A. S. Butement, R. K. Eisenhitz, A. W. Foster, P. G. Law, R. W. B. Nurse, W. H. Reynolds, E. E. Schneider, C. J. Smith, M. Stephenson, Tolansky, S. Weintraub.

**Associates:** L. Bagnall, B. M. Bellman, C. J. Brown, M. Brown, J. Burfoot, R. Butler, J. C. E. Button, L. Cardell, B. A. M. Caspersz, P. Cliffe, J. Collins, R. D. Connor, H. Coupland, H. W. Duckworth, P. G. Dutt, D. A. Dyer, Francis, J. Franks, I. A. Gatenby, E. George, S. R. Govindarajan, H. W. Grimwood, G. M. Habberjam, J. S. Halliday, J. W. S. Hearle, O. S. Heavens, T. L. Howson, O. Isard, A. H. Jarrett, R. B. Johnson, J. P. Jones, J. J. Knight, E. T. Linacre, W. C. Lister, T. A. Lucas, J. M. Macaulay, R. C. H. MacCormac, J. W. Menter, P. A. Michaels, E. Miles, D. Mowatt, G. A. J. Orchard, L. G. Parry, E. Pattinson, J. M. Peskett, D. C. Phillips, A. W. Pratt, B. Rajchenbaum, J. D. Ranade, F. R. Reavell, D. A. Richardson, E. Rule, J. G. Santesmases, A. A. Shepherd, R. W. Smith, E. Stanley, R. M. Tennent, H. Tunley,

B. M. Wheatley, P. S. Williams, P. Wootton, K. H. R. Wright, B. Yates.

One hundred and eleven Graduates, twenty-nine Subscribers and seventy-three Students were also elected.

**X-ray Analysis Equipment.** The Equipment Sub-Committee of the X-ray Analysis Group of The Institute of Physics makes the following announcements regarding the work of its Panels: (1) A user specification for high-temperature X-ray powder cameras has been prepared by the Powder Camera Panel. (2) A user specification for a microdensitometer suitable for X-ray powder photographs has been drawn up by the Photometer Panel.

Copies of these specifications may be obtained (free of charge) from the Secretary, The Institute of Physics, 47 Belgrave Square, London, S.W.1.

**Conference on the Physics of Lubrication.** A conference arranged jointly by the Manchester and District Branch of The Institute of Physics and the British Rheologists' Club will be held in the University of Manchester from 29 June to 1 July next, on "The Physics of Lubrication." The general arrangements will be similar to those for the conferences arranged by the branch in previous years. It is intended to

devote sessions to fluid film lubrication, boundary and extreme pressure lubrication, and the physics of lubricants.

Suggestions for papers will be welcomed and should be sent, as soon as possible, to the Conference Secretary, Mr. D. A. Crooks, Imperial Chemical Industries, Ltd., Dyestuffs Division, Hexagon House, Manchester, from whom further details may be obtained.

**Educational Booklets on Thermionic Valves.** An interesting and well-produced series of educational booklets is being issued by The Edison Swan Electric Co. Ltd., under the general title "Electrons." The series deals with diodes, triodes, screened grids, pentodes, beam tetrodes, frequency changers and multiple diodes. Each booklet is written in concise and attractive English and is well produced and illustrated with simple two-coloured diagrams which are a model of what can be achieved with a little imagination. It is an excellent thing for all those concerned with applied physics to know of the useful help which industrial firms are increasingly giving to students and teachers by providing educational literature of this nature. Copies of the booklets, of which the first two have been published, are available free to Education Authorities, Radio Societies and other bona fide bodies.

**Modern Permanent Magnets.** We have received a copy of an extravagantly produced 68-page volume entitled *Permanent Magnets*—a Jessop-Saville Technical Publication. This is a contribution to information on the history, technical criteria and production of permanent magnets, to

which these firms have contributed much. The volume is remarkable as an example of the interest that can be aroused by exploiting modern methods of lay-out and printing; for example, the graphs, figures and tables are produced in three- or four-colour printing and the whole style is reminiscent of the modern pictorial magazines. Such a style may be a good thing in a world flooded with scientific literature and requiring something outstanding to attract the attention of the reader, but it must be very expensive. When a firm is competing for the sale of its products in the world's markets of to-day such an expense may be justifiable but how far it should or could be incurred by publishers of educational books is a debatable point. Our own view is that most text-books of physics could be made a good deal more attractive to the student without an appreciable increase in price, but there is no need to dress up such a fascinating subject in the way that advertisers may find it necessary to do.

The book is intended primarily for the designer who needs permanent magnets in his products, but the enterprising teacher will find it refreshing and helpful. The full-page frontispiece of this volume—a remarkable picture showing the iron atom with its 26 orbital electrons and indicating the paths of the assumed missing electrons—and other illustrations will help him to explain the modern theory of magnetism.

We understand that this is the first of a series these firms intend to produce and we look forward to receiving copies of later ones; we hope, too, that others will also find it possible to include some fundamental physics in their technical literature.

## Journal of Scientific Instruments

### Contents of the April Issue

#### SPECIAL ARTICLE

Radioactivity Surveying and Monitoring Instruments. By D. Taylor.

#### ORIGINAL CONTRIBUTIONS

Considerations in the Design of Demountable X-ray Diffraction Tubes. By R. I. Garrod.

Variation of Sensitivity with Speed in an Eddy-Current Torsionmeter. By D. C. Johnson.

A Proportional Reset Furnace Controller for Temperatures up to 1600° C. By R. W. B. Nurse and J. H. Welch.

An Improved Nuclear Disintegration Camera. By J. H. Sanders.

The Preparation of Metal Single Crystals for Expansion Measurements. By B. G. Childs.

A Method of Decreasing the effect of Earth Admittances in a.c. Bridges. By G. H. Rayner and R. W. Willmer.

A Method for the Summation of Irregular Linear Movements. By P. W. Roberts.

Photographic Materials for use in the Ultra-violet. By R. D. B. Fraser.

#### NOTES AND NEWS

#### *New Instruments, Materials and Tools*

Optical Pyrometer—Precision Drawn Wave Guides—Recording Camera—Photoelectric Spectrophotometer—Two-Circle Optical Goniometer—Direct-Reading Electronic Clock and Timer—Wheatstone Bridge Engine Speed Indicator.

#### *New Books*

Radio Engineering—The Technique of Radio Design.

## British Journal of Applied Physics

### Original Contributions accepted for publication in future issues of this Journal

Diffusion of Moisture through Flax Seed. By D. E. Samuels and B. H. Tait.

Methods of Storing and Handling Liquefied Gases. By A. Croft and G. O. Jones.

Electrical Conductivity of Oxide Cathode Coatings. By D. A. Wright.

Soft Soda Glass to transmit Bactericidal Radiation. By J. E. Stanworth.

A method of locking Oscillators in Integral and Non-Integral Ratios. By E. A. G. Shaw.

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**EDITORIAL MATTER.** Communications concerning editorial matter should be addressed to the Editor, The Institute of Physics, 47 Belgrave Square, London, S.W.1. (Telephone: Sloane 9806.) Prospective authors are invited to prepare their scripts in accordance with the *Notes on the Preparation of Contributions* which will be sent gratis on request.

**ADVERTISEMENTS.** Communications concerning advertisements should be addressed to the agents, Messrs. Walter Judd Ltd., 47 Gresham Street, London, E.C.2. (Telephone: Monarch 7644.)

**SUBSCRIPTION RATES.** A new volume commences each January, and annual subscriptions will only be accepted for complete volumes of the monthly parts (January–December). The charge is £3 per volume (\$8.50 U.S.A.), including index (post paid), payable in advance. Single parts, so far as available, may be purchased at 6s. each (85c. U.S.A.), post paid, cash with order. Orders should be sent to The Institute of Physics, 47 Belgrave Square, London, S.W.1, or to any Bookseller.



## ORIGINAL CONTRIBUTIONS

## The Rheology of Unseasoned Linoleum

By A. G. WARD, M.A., F.Inst.P., Building Research Station, Garston, Watford\*

*[Paper first received 16 December, 1949, and in final form 18 January, 1950]*

An extrusion method for the measurement of the flow properties of unseasoned linoleum is described. The method is suitable either for the study of absolute flow properties, or for routine testing, of unseasoned linoleum, and examples of its application are given. The significance of the method in relation to the manufacture of linoleum is discussed, particularly in relation to the effects of combined hydrostatic pressure and shearing stress as in the calendering process.

The essential constituents of linoleum, as normally manufactured, are a binder or "cement" containing oxidized linseed oil, gums and rosin, and fillers made up from ground cork, wood flour, and pigments (including whitening). The treatment of the binder<sup>(1)</sup> produces a stiff, rubbery gel, into which the filler is incorporated in the manufacturing process. The relative proportions of the different types of filler, and the filler/binder ratio are varied according to the type of linoleum required. The rheological properties of the binder, both alone and when partially mixed with the filler, are of great importance in the various stages of the incorporation process, but the problems involved are so complex that no serious attempt has yet been made to study them. The present work was undertaken to devise a method of test for the incorporated linoleum which would enable the behaviour of the material during calendering to be controlled.

The final stage of the incorporation involves the extrusion of the material through a die plate. The extruded material, in which the binder has been distributed so that the filler particles are coated with it, then passes to two small rolls, forming a sheet in the nip of the rolls. It is then torn off the roll to which it is adhering, by a spiked roller, to give a crumbly powder resembling bread crumbs. This material drops on to the main calendering rolls which are of massive construction. It is necessary to ensure that the sheet adheres to one roll, and this is achieved by running one roll about 100° F (55·5° C) cooler than the other, since the adhesion increases rapidly as the temperature is lowered. The colder roll is run slightly faster than the hot roll. The sheet is then detached from the cold roll, and either calendered direct on to the backing or used in the production of inlaid, jaspe or marble patterned linoleum. In the latter two processes, portions of sheets of different colour are used in conjunction with a further set of calendering rolls to produce the required pattern. The flow properties of the sheets will influence the pattern obtained. The final product is then "seasoned" in large ovens, where the material becomes harder and less plastic.

In sheet formation it would appear that the rolls first compress the powder, with some shearing action, until a continuous sheet is formed, which is then compressed and sheared in passing through the nip of the rolls. The deformation is almost entirely plastic, elastic recovery being extremely small. The rolls therefore subject the material to a combined stress, made up of hydrostatic pressure and shearing stress. The same system of combined stresses is present in the formation of jaspe and marble patterns. A method of test must therefore also subject the material to a similar stress system if the results are to be compared with practical calendering.

Before describing the method of test, consideration will be given to the formulation of the linoleum in a way which will enable the influence of composition on flow properties to be studied systematically.

## FORMULATION ON A VOLUME BASIS

Works practice necessarily means that the composition of linoleum is expressed on a weight percentage basis. In considering the influence of different fillers on flow properties it is necessary to express compositions on a volume basis, since, if specific effects are absent, and the fillers can be regarded as made of rigid particles, an interchange of fillers of similar particle size distribution would leave the flow properties unchanged. Where the particle size distributions differ, the question of packing will also be relevant, but it is still necessary in the first instance to use volume percentage compositions.

The real densities of the component materials used in linoleum were, therefore, determined. Two methods were used for the measurement of real density. The first was the vacuum density bottle method. A 50 ml density bottle was used, with kerosene as the liquid for pigment, wood flour and ground cork, and water for the binder. Sufficient powder was added to the density bottle to occupy about 25% of the volume, whereas for the binder a block was cut into pieces and as many as possible of these were packed into the bottle. The bottles were evacuated after weighing, and the appropriate liquid run in while the vacuum was maintained. Evacuation was then continued for a period. The bottles were then removed to a 25° C thermostat, and topped up

\* Now at—The British Gelatine and Glue Research Association, 1/4 Dalmeny Avenue, London, N.7.

in the usual way. The densities were then calculated from the densities of the liquids and the volumes of the bottles. Errors are likely to arise in this method from failure to fill capillary spaces, or from totally enclosed pores. A small fraction of the ground cork floated on the liquid, indicating one or other of these sources of error. The density required in calculation of the composition should exclude errors from capillaries, but since the closed pores will also be inaccessible to the linseed oil binder, no allowance should be made for them.

A second method of measurement was used which ensured the filling of capillaries but not enclosed pores, with air as the medium. A density bottle was filled with powder and connected to an inverted burette. By adjusting the height of a mercury reservoir attached to the lower end of the burette, the air was subjected to a number of different pressures, measured by the addition of atmospheric pressure to the difference in mercury levels in the burette and a tube of equal diameter open to the atmosphere. The volumes were read on the burette. Then from Boyle's Law the free space in the density bottle was obtained. From this the density of the powder was readily calculated. There was no advantage in attempting to increase the accuracy of the measurements beyond about 1% owing to the variability in the materials. The results are given in Table 1.

Table 1. *Density of the constituents of certain linoleum samples*

		Density bottle method		Dead space method		Value used
		Density	Mean	Density	Mean	
Binder	(1)*	1.085		1.079		
		1.088	1.087	1.066	1.072	1.08
Wood flour	(1)	1.396		1.423		
	(2)	1.418		1.450		
	(3)			1.432		
	(4)		1.407	1.406	1.428	1.41
Ground cork	(1)	1.249		1.234		
	(2)	1.324		1.234		
	(3)			1.268		
	(4)		1.287	1.267	1.251	1.25
Pigment	(1)	2.811		2.788		
	(2)	2.811		2.803		
	(3)			2.787		
	(4)		2.811	2.833	2.803	2.81

\* The numbers refer to duplicate experiments on material from the same batch.

It will be seen that the pigment gave very good accuracy by both methods, and that the results agreed. The only doubtful values were those for ground cork. Since some of the ground cork floated on the liquid in the density bottle method, the "dead space" figures have been used for the final value.

The ground cork showed a wide range of particle size grading, from a diameter of about 0.03 in to 0.003 in, with a somewhat similar grading for the wood flour. The pigment and whiting were finely divided, only 2-3% being retained on the 200-mesh sieve.

Preliminary experiments were performed in which the powder from the scratcher rolls was formed into cylindrical pellets using pelleting moulds. Both mould and powder were heated before the powder was consolidated under pressure.

It was immediately evident that, although both temperature and pressure used were equal to, or in excess of that found in calendaring, the pellets were very poorly consolidated. On compressing the pellets, failure in shear occurred after quite small compressions, confirming the lack of cohesion. The bulk density of the pellets was not significantly different from that of the calendared sheet material, but it was clear that the individual particles from the scratcher rolls had failed to combine in the formation of pellets.

The manufacture of coherent linoleum sheet can therefore be considered to require shear in addition to hydrostatic pressure. The explanation is probably partly the need to expose fresh tacky surfaces in the binder and also to allow entrapped air to escape without forming channels, which would be sources of weakness.

A few experiments were performed in which a small disk of the sheet linoleum was compressed to a fraction of its thickness in the parallel plate plastometer. It was observed that the material at the centre of the disk remained coherent even under the large shear which had occurred, whereas the material at the edge had cracked. This can be explained by the presence of hydrostatic pressure in the central region of the pellet, which is a consequence of the frictional forces between the pellet surface and the plastometer plate.<sup>(2)</sup> At the outer boundary, the hydrostatic pressure was small and rupture took place. It would therefore appear that, even at high temperatures, continuous flow of consolidated linoleum is only possible if some hydrostatic pressure is present to prevent rupture.

Hydrostatic pressure is necessary not only to simulate the stresses of calendaring but also to enable continuous shearing to occur. The only readily available experimental methods giving the appropriate stress system are extrusion and compression of thin disks. The latter is quite unsuited to the investigation of a new material, since the results cannot be analysed in terms of stress and strain. Extrusion, on the other hand, is both convenient and, in certain circumstances, capable of analysis. Calendaring can be regarded as two-dimensional extrusion, to a first approximation, so that the relation between the experimental method and the industrial operation is close.

#### THE EXTRUSION METHOD

The general arrangement of the extrusion apparatus is shown in Fig. 1. The cylindrical chamber was 1½ in diameter and 5 in long, and was surrounded by a jacket



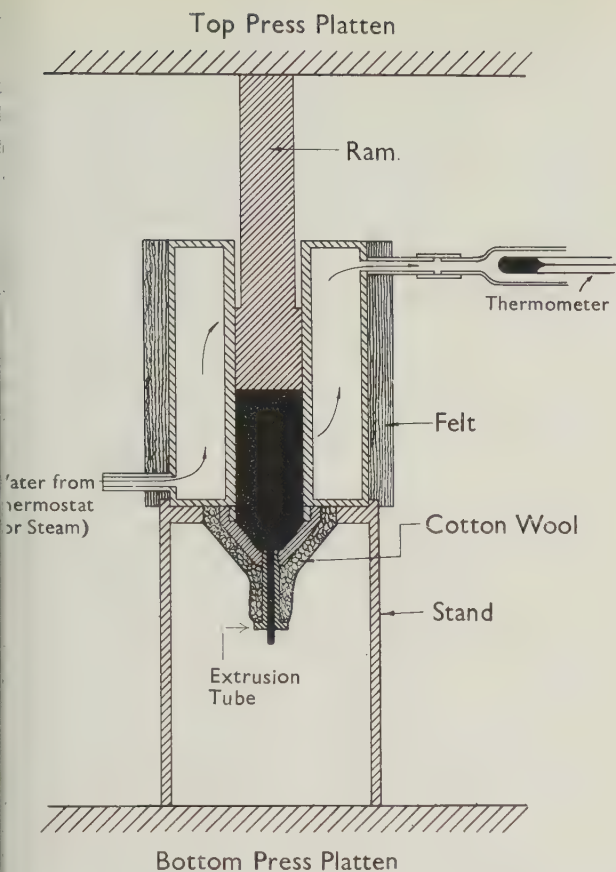


Fig. 1. Diagram of extrusion apparatus

through which water could be circulated from a thermostat. The extrusion tubes were fitted with small heater sleeves containing resistance wire, so that they could be heated. A thermocouple was inserted in a hole 1 in long in the wall of the tube, parallel to the axis, and the current through the heater was controlled to maintain the temperature of the tube at the same figure as the extrusion chamber. The ram was a good fit in the chamber, the actual surface in contact being  $2\frac{1}{4}$  in long. The lead-in to the cylindrical dies was cone-shaped, with semi-vertical angle  $45^\circ$ . The tubes were screwed into the lead-in and consisted of a set of nine, made by combining three lengths with three diameters as set out in Table 2.

Table 2. Dimensions of the nine tubes

Tube number	Radius in cm	Length in cm
1.1	0.323	10.16
1.2	0.251	10.20
1.3	0.159	10.15
2.1	0.320	5.11
2.2	0.254	5.11
2.3	0.166	5.10
3.1	0.326	2.56
3.2	0.240	2.61
3.3	0.160	2.54

The apparatus was supported on a stand, giving free access to the end of the tube, so that the rate of extrusion could be determined by cutting off the length extruded in a given time. Pressure was applied by means of a hydraulic press, and it was measured in the first instance by the reading of the pressure gauge of the press. This gauge pressure was related to the force exerted on the ram by calibration of the gauge with a standard ring gauge between the press platens. Frictional forces on the ram were estimated at the conclusion of each experiment by removing the lead-in and measuring the pressure to force the plug of linoleum out of the chamber.

Sheet linoleum was used in the extrusion experiments in preference to powder, in order to ease filling the extrusion chamber. Disks were cut from the sheets, and these were then preheated for a specified period, transferred to the heated chamber, and then the final temperature adjustments were made. A total heating period of  $2\frac{1}{2}$  hr was allowed.

It was observed that the properties of the linoleum changed appreciably during a lengthy experiment so that in comparing two materials, a rigid time-table was observed. Protracted experiments on any one material were avoided. For each tube the procedure was to increase the applied pressure until extrusion occurred at a measureable rate. The rate was measured at several values of the applied pressure up to the maximum figure at which the rate could still be measured. One of the measurements at a low rate of extrusion was then repeated to verify the absence of thixotropic effects or some defect in the apparatus. The lengths cut off were weighed and converted to the volume rate of extrusion  $Q$  ml/sec, using the measured density.

#### ABSOLUTE FLOW PROPERTIES

Measurements of extrusion through the full set of nine tubes were made for one material, in order to see whether the absolute flow properties of linoleum could be determined in C.G.S. units. The material used contained 37% by weight of binder (43% by volume) with a filler of high ground cork content and low pigment content. All measurements were made at  $100^\circ\text{C}$ , this temperature being chosen to increase the number of measurements which could be made with the finer tubes. In order to avoid the progressive hardening of the material at  $100^\circ\text{C}$ , separate experiments were performed for each set of three tubes of the same length. By retaining the same order of use of the tubes of different diameter in the three experiments, the effect of ageing on the results was almost completely eliminated. The method of analysis of the results has been fully described by Ward and Westbrook,<sup>(3)</sup> and only a brief description will be given here.

If a material flows through a tube, with no slip at the wall, with streamline flow, and with no thixotropic or similar effects, then a plot of  $P^*$  against  $Q^*$  gives a single

curve for all tube lengths and diameters.  $P^*$  and  $Q^*$  are defined as:

$$P^* = Pr(6.89 \times 10^4)/2L \quad (1)$$

$$Q^* = 4Q/\pi r^3 \quad (2)$$

$P$  is the applied pressure in lb/in<sup>2</sup>,  $r$  is the tube radius,  $L$  is the tube length, and the numerical factor in equation (1) is to convert lb/in<sup>2</sup> into dynes/cm<sup>2</sup>. The relation between shearing stress  $\tau$  and rate of shear,  $D$ , may then be obtained from the single curve of  $P^*$  against  $Q^*$ ,<sup>(4)</sup> using the relations:

$$\tau = P^* \quad (3)$$

$$D = \frac{1}{4}(3Q^* + \tau dQ^*/d\tau) \quad (4)$$

The measured pressure in the experiments was, however, made up of two parts,  $P$  the pressure to force the material through the tube and  $\Delta P$  the pressure to bring it from the chamber into the tube. For materials such as linoleum, the latter may form an appreciable proportion of the total pressure.  $\Delta P$  can be found by taking two tubes of length  $L_1$  and  $L_2$  of the same nominal diameter, and plotting  $Q^*$  against  $P$ . If the diameters of the tubes are identical, then  $Q$  can be used instead of  $Q^*$ . Then, from the graph, the values of  $P_1$  and  $P_2$  are taken, being the pressures for the two tubes for the same value of  $Q^*$ . The difference  $P_2 - P_1$  is the pressure necessary to force the material through a tube of length  $L_2 - L_1$ . The pressure necessary for the full-length  $L_2$  is then:

$$(P_2 - P_1)L_2/(L_2 - L_1)$$

This quantity can be subtracted from the pressure  $P_2$  to give  $\Delta P$ . If all the necessary conditions are fulfilled the value of  $\Delta P$  should be independent of which pair of

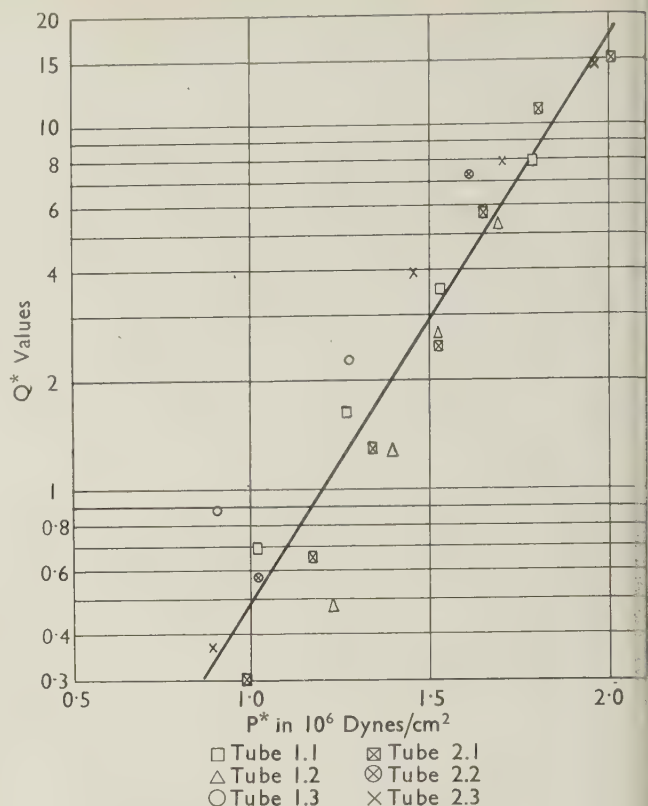


Fig. 3.  $P^* - \log Q^*$  at 100°C

tubes of the same nominal diameter is taken. This was not found to be so, but the discrepancies were not systematic and probably resulted from errors in the values of  $P_1$  and  $P_2$  which, owing to the form of the flow curves for linoleum, would produce substantial errors in the value of  $\Delta P$ . In order to arrive at the best values for  $P$ , the values of  $\Delta P$  used for the long tubes were derived from results using long and medium tubes; for the short tubes, results from short and medium tubes were used; and for the medium tubes the mean value was used. It was found that the end correction for the short tubes formed so large a proportion that it was not possible to use them, but for both medium and long tubes the end correction was subtracted from the pressure to give the values of  $P$ . From this  $P^*$  was calculated.

Fig. 2 gives the graph of  $P^*$  against  $Q^*$  for the six tubes. Taking into account the errors involved in the derivation of  $\Delta P$ , the points may be considered to fall on a unique curve, so that it would appear that the necessary conditions of flow are fulfilled in the passage of the linoleum through the tubes at 100°C. It is very unlikely that this result would have been obtained had there been appreciable slip at the tube walls.

In Fig. 3 the results are plotted as  $\log Q^*$  against  $P^*$ . The best straight line was fitted to the points in the usual way. The equation for the line is:

$$\log_{10}(Q^*) = 0.319 + 1.53(P^* - 1.41) \quad (5)$$

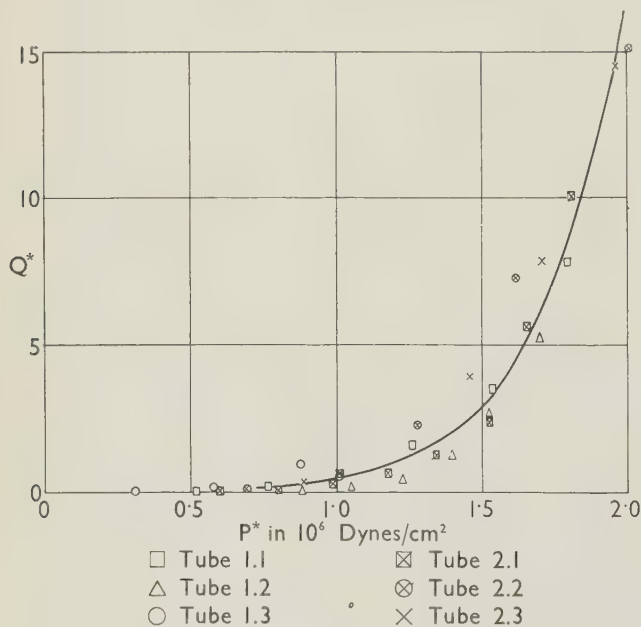


Fig. 2.  $P^* - Q^*$  graph at 100°C



Using this line, values of  $Q^*$  were obtained for selected values of  $P^*$  and these were plotted on Fig. 2 to give the line shown there. It should be noted that units of  $10^6$  dynes/cm<sup>2</sup> were used throughout.

In order to obtain the absolute flow relation between the rate of shear and the shearing stress, equation (4) must be used. This can be obtained graphically from the  $P^*Q^*$  diagram, but since equation (5) gives an analytical expression the procedure is simplified.

In general:

$$\log Q^* = A + B(\tau - \tau_0) \quad (6)$$

$$dQ^*/d\tau = BQ^*$$

therefore, from (4),

$$D = (Q^*/4)(3 + BP^*) \quad (7)$$

The value of  $B$  from equation (5) is  $2.303 \times 1.53$ , and using this the values of  $D$  for different values of  $(= P^*)$  were obtained. The results are shown in Fig. 4. If we

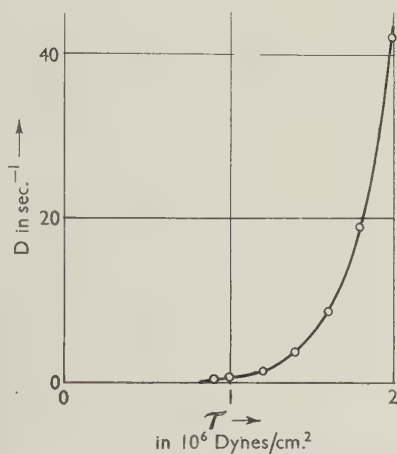


Fig. 4.  $D - \tau$  diagram at 100°C

substitute from equation (6) in equation (7) we obtain the explicit relation between  $D$  and  $\tau$ .

$$D = (A/4) \exp [B(\tau - \tau_0)] (3 + B\tau) \quad (8)$$

this would be required to enable the stress to be calculated in an operation in which the rate of shear is known. In many instances, however, the rate of shear will not be easy to obtain since the flow may be extremely complex.

Although the absolute method of analysis has been applied satisfactorily to the results for this sample of linoleum at 100°C, it does not follow that it will be applicable to all types of linoleum or all temperatures. It would appear probable that the onset of slip at the tube wall would be the most likely source of difficulty. If this does occur, the analysis given should rapidly detect it.

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## STUDIES OF THE FORM OF THE FLOW THROUGH TUBES

Two methods were tried in an attempt to see what was occurring during the extrusion. In the first, two samples of unseasoned linoleum were used, the colours of which were appreciably different. The cylinder was charged with alternate disks of the two colours at 100°C, and then extrusions were conducted at different extrusion pressures. Lengths of the extruded material were cut off and then cut open parallel to the length through the axis. The pattern produced was clearly visible and showed at once that very considerable shear was occurring during the extrusion. The softer of the two materials had been brought almost to a point at the tip, whereas the harder material had come through with more of a plug shape, surrounded by the next layer of soft material. The difference is that which would be expected since the "yield value" for the harder material is higher than that of the softer material.

In the second set of experiments, the disk surfaces of one sample were dyed with a mixture of aqueous "brilliant green" and "crystal violet." Extrusions were conducted at 100°C as before. The same general results were obtained and Fig. 5 shows scale drawings of the positions of the boundaries between the disks, at different extrusion pressures. With the exception of the result at 1,600 lb/in<sup>2</sup> it is clear that the sharpness of the point increases as the stress is raised. This would be expected on theoretical grounds. Fig. 5 also shows a section of the residue of the dyed material remaining in the chamber and entry to the extrusion tube, showing clearly how the disks were deformed. These results broadly confirm the results of the absolute measurements that slip, although not completely excluded, is insufficient to modify appreciably the shear undergone by the linoleum in its

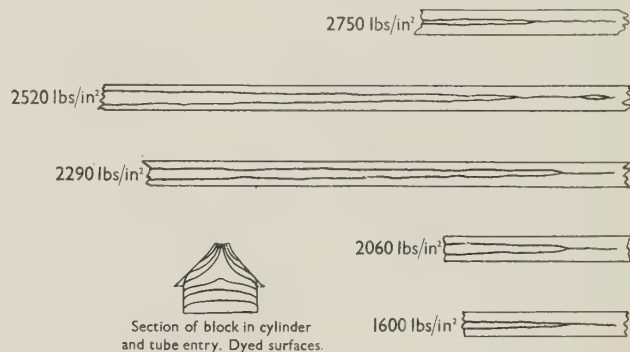


Fig. 5. Extrusion patterns. Dyed surfaces of linoleum samples

passage through the tube. In addition the results obtained with the two samples of different colour, which also differed in hardness, may have some significance in relation to the production of jasper and marbled effects.

\*

## TEMPERATURE DEPENDENCE OF FLOW PROPERTIES

The sample used in these measurements had 35.8% of binder (43% by volume) with a higher pigment content than the sample used in the absolute measurements. The results are shown in Fig. 6, where the log (extrusion

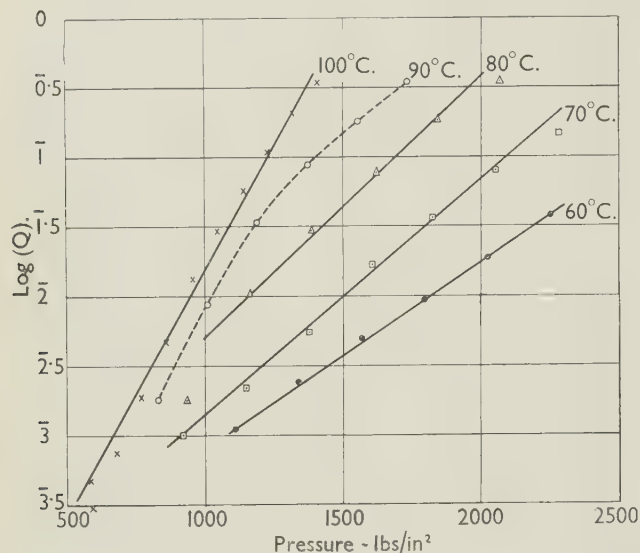


Fig. 6. Relation between log (extrusion rate) and pressure, 60°–100° C

rate) is shown as a function of the applied pressure. The precision of the measurements, which were only carried out for one tube, was insufficient to enable the quantitative variation of the flow with temperature to be deduced.

## DISCUSSION

The extrusion method has, in the experiments which have been described, shown that it can provide useful results for unseasoned linoleum. It can be used, at least for certain compositions, to measure the absolute flow properties. It appears both from the use of the  $P^* - Q^*$  graph and from actual study of flow contours that slip is not of importance. The form of the dependence of rate of flow on applied stress means that the rate is very sensitive to the stress, or conversely, fairly wide variations in rate of flow produce only small changes in the stress. It is probable that these facts render the material suitable

for calendering. In addition there is a fairly well-defined yield stress.

By using only one tube, the apparatus can be made into a convenient routine instrument and the results which are obtained in this way show good agreement with results in practice on the calendering rolls. By this means the effect of variation of the constituents can be easily followed, and also the temperature dependence.

Elastic recovery, according to measurements of the diameter of the extruded sample, was negligible under the conditions of the extrusion experiments. In fact any expansion which did occur was masked by the subsequent thermal contraction of the sealed pockets of air in the material. The percentage by volume of air in the sheet samples studied in this investigation varied from 10.3% to 4.5%, but the major part of this was driven out in the extrusion process. At the higher rates of extrusion, it was observed that the surface of the extruded material became feathered and torn, whereas at the lower rates it was highly polished. It may be that this effect results from the elastic forces having insufficient time to relax during passage through the tube.

Preliminary experiments suggest that the packing of the filler material alone may provide a useful guide to flow properties. Such investigations require, however, to be linked with systematic measurements of flow properties, starting with the gel binder alone and then adding the constituents one by one.

## ACKNOWLEDGEMENTS

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# Method of Selection of Kilovoltage on X-Ray Sets, and its effect on X-Ray Output

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In X-ray sets employing an auto-transformer and variable resistance as rough and fine voltage controls, in the primary circuit, there are a number of auto-transformer tapings, which each with a suitable value of resistance, will give the correct reading on the voltmeter. Though the tube current is adjusted to its correct reading in the normal manner in every case, the output quality and quantity will alter with transformer tapping. The effect has been investigated on several sets and is also discussed from a theoretical point of view. Recommendations are made for minimizing the effect.

Amongst methods used to control the kilovoltage on X-ray sets, a common one is to use an auto-transformer as a rough control and a series adjustable resistance as a fine control, both in the primary circuit (Fig. 1). The

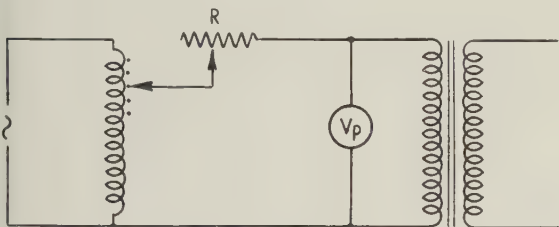


Fig. 1. Kilovoltage control

secondary of the transformer may be connected directly to a self-rectifying X-ray tube, or to a rectifying circuit. The meter  $V_p$  is used to set the kilovoltage and although it is across the primary of the transformer, it is usually calibrated to read kilovoltage across the secondary.

It will be seen that it is possible to select any of a number of auto-transformer tapings and by suitable adjustment of  $R$ , to set the chosen voltmeter reading. The chosen tube current is maintained in the normal manner, using an adjustable resistance or inductance<sup>(1)</sup> in series with the filament winding.

It has been found that, in general, the X-ray output varies both in quantity and quality with the auto-transformer tapping selected and that the variation may in certain circumstances be of the order of 50%. It should be stressed that this is not the well-known effect in which the X-ray output varies because of the alteration of kilovoltage when the tube current is altered, or vice versa. The effect now under consideration alters the X-ray output with both voltmeter and milliammeter set correctly; it has been noted before by Stewart,<sup>(2)</sup> but no general investigation has been found in the literature. Since it is possible for quite serious errors to enter into radiotherapy on account of this effect, while the operator has every reason to believe that the set is functioning correctly, it was felt that publication of an investigation would be worth while.

## THEORETICAL DISCUSSION

In discussing the effect of the running conditions on the quality and quantity of output, five main types of sets have to be considered and three well-known relationships governing output.

The five main types of set are:

- (a) Self-rectification.
- (b) Unsmoothed half-wave rectification.
- (c) Unsmoothed full-wave rectification.
- (d) Smoothed half-wave rectification.
- (e) Smoothed full-wave rectification.

The governing relationships are:

- (1) An increase in voltage applied to the tube increases the output in an approximately square law relationship.
- (2) An increase in voltage applied to the tube increases the H.V.L.
- (3) For a given voltage, the X-ray intensity is directly proportional to the tube current, within certain limits.

Now exactly what the voltmeter  $V_p$  reads, will depend on the type employed. A moving-iron instrument reading r.m.s. values would be usual, but in all cases it can be said to read a "time-average" of the primary voltage.

In self-rectification of type (a), this voltage will include that for both an off-load and on-load half-cycle. If  $R = 0$  (Fig. 1), the primary voltage will be equal to the auto-transformer output during both these half-cycles and will be, say,  $V_0 \sin \omega t$ . If, however,  $R \neq 0$ , during the off-load half-cycle, the voltage will be greater than  $V_0 \sin \omega t$ , since the auto-transformer output is increased and the secondary current zero and hence the primary current very small. It therefore follows that during the on-load half-cycle, the "time-average" primary voltage will be less than that for  $V_0 \sin \omega t$ , since the voltmeter reading over the whole cycle is adjusted to read the same whatever the value of  $R$ ; this will in general lead to a decrease in output and a softening of the radiation. This is the most important factor, but a change in voltage wave-form may also be expected. Owing to the non-

ohmic characteristics of the X-ray tube, as saturation current is approached, the secondary and therefore the primary current would be expected to increase less rapidly than the sinusoidal voltage. Hence the voltage drop across  $R$  would also increase less rapidly than the sinusoidal voltage, as it represents the difference between a sinusoidal wave-form and the drop across  $R$  and hence  $V_p$  would be expected to be more sharply peaked than for a sinusoidal wave-form.

The above arguments apply also to half-wave rectification of type (b), but here there is an additional factor in that the secondary voltage distribution between rectifiers and X-ray tube will depend on their relative impedances, so that a distinction must be made between transformer secondary and tube kilovoltages. Since it is usual to run the tube under saturation conditions when its impedance increases with kilovoltage and to run the rectifiers at voltages below those required for saturation, when the impedance is more nearly constant, any decrease in secondary kilovoltage will lead to a less sharply peaked tube voltage wave-form and vice versa. To summarize, the fall in peak kilovoltage when  $R \neq 0$  compared with that when  $R = 0$  will lead to a flattening of the tube voltage wave-form, whereas the presence of resistance in the primary will by itself lead to a peaking of the whole secondary voltage wave-form and no generalization can be made about the net effect on the tube voltage. Apart from the wave-form changes, however, there will be the drop in kilovoltage over the half-cycle, as described above, and hence a decrease in output and a softening of the radiation can in general be expected.

In the case of unsmoothed full-wave rectification [type (c)], both half-cycles are on load and the primary voltage "time-average" will be the same for  $R = 0$  and  $R \neq 0$ . However, the argument regarding the peaking of the secondary voltage wave-form in the cases described above should apply and hence the peak kilovoltage should be slightly raised when  $R \neq 0$ . This in turn should lead to a further peaking of the tube wave-form, because of the relative impedances of tube and rectifiers as in type of set (b). Thus a greater and harder output would be expected when  $R \neq 0$ , compared with that for  $R = 0$ , for full-wave unsmoothed rectification.

In types of sets (d) and (e), if the smoothing and regulation were perfect, the load would be constant throughout the cycle. This implies a large capacity in the smoothing circuit, with a nearly constant voltage. Hence the on- and off-load conditions would be nearly the same and any effect due to  $R$  would be small. In practice smoothing and regulation will not be perfect and the results will be intermediate between those described for perfect and for no smoothing.

So far, the effects of changes in tube kilovoltage only have been considered, but, except where the tube kilovoltage is that for saturation current throughout the cycle, changes in the former will cause changes in tube current. In contradistinction to changes in voltage,

where the peak values are most important, the lower voltage region of the cycle will have most effect on tube current, because of the usual saturation conditions under which the tube is run. If, when  $R \neq 0$ , the voltage is greater than when  $R = 0$ , in these lower voltage regions, the overall tube current will increase and in order to restore it to the chosen value, the filament heating current will have to be lowered. This will lead to a lower saturation current and a consequent lowering of the output. Though it is difficult to predict just when such changes will occur, without detailed knowledge of tube and rectifier characteristics and circuit constants, these wave-form changes in tube current have been found in practice. However, because of the linear relationship between current and output, compared with the square law relationship between voltage and output, the current wave-form changes may be expected to have smaller effects than those due to changes in voltage wave-form.

#### EXPERIMENTAL

The effect of a resistance in the primary circuit was investigated in the following sets:

- (1) Maximar 220-kV Deep Therapy Set. Self-rectification.
- (2) Dean 150-kV Superficial Set. Half-wave rectification, unsmoothed.
- (3) Watson Diagnostic Set. Full-wave rectification, unsmoothed. (Specially modified.)
- (4) Watson Diagnostic Set. Half-wave rectification, unsmoothed. (Specially modified.)
- (5) Metalix Contact Set, 50-kV. Half-wave rectification, smoothed. (Specially modified.)

#### (1) MAXIMAR 220-KV DEEP THERAPY SET. SELF-RECTIFICATION

A simplified circuit of this set is shown in Fig. 2. It will be seen that this set has a suppressor in the primary

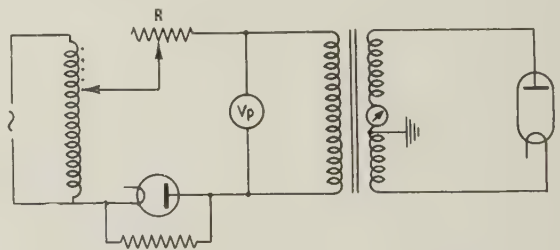


Fig. 2. Maximar 220-kV deep therapy set (simplified circuit)

circuit to reduce the h.t. transformer voltage during the off-load half-cycle. This will decrease the difference between on- and off-load half-cycles and make the behaviour of the set intermediate between that for half- and full-wave rectification (unsmoothed).

The auto-transformer has stud positions numbered 1 to 52, the control of which is on the front panel, number 52 corresponding to the highest voltage output. The



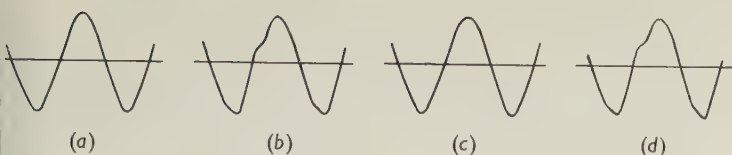


Fig. 3. Maximar primary voltage wave-forms

(a) Stud 23, off load.  
(b) Stud 23, on load.

(c) Stud 16, off load.  
(d) Stud 16, on load.

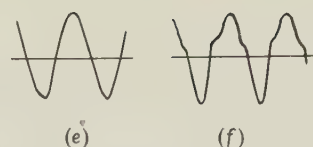


Fig. 4. Maximar primary voltage wave-forms

(e) R small, on load.  
(f) R large, on load.

adjustable resistance has numbered positions 1 to 23, the latter corresponding to the lowest value of resistance.

Output measurements were taken in air, with various filtrations, using a Victoreen and with the auto-transformer adjusted so that the resistance control had to be either at stud 23 or at stud 16 in order to give a kilovoltage reading of 220. The tube current was controlled manually at 15 mA.

The results are shown in Table 1. The equivalent kilovolts were obtained from a graph of the relation between H.V.L. and equivalent kilovolts published by Mayneord and Lamerton.<sup>(3)</sup> It is evident from these results that the output is reduced both quantitatively and qualitatively by the introduction of R.

Fig. 3 shows the primary voltage wave-forms. It will be seen that while there is some distortion from a sine wave towards peakiness when R is low, it is more marked when R is increased. The differences are, however, rather small. But in order to clarify the change in primary voltage wave-form, additional photographs were taken, with a much larger value of R in the primary circuit. These are shown in Fig. 4, where it is evident that the peak of the curve is narrower when R is large, than when this is not the case, as would be expected from the theoretical discussion.

It is difficult to see the precise results of wave-form changes in Table 1, where it must be borne in mind that all figures apply not to the total output but to more or less heavily filtered radiation. A striking feature of the results is the way in which the percentage decrease in output when  $R \neq 0$ , compared with that when  $R = 0$ , increases with filtration, but this is to be expected simply because the peak kilovoltage is higher and hence the radiation harder in the latter case.

These results may be taken as applying to the worst case that would occur in practice. Stud sixteen corresponds to seven steps in resistance, whereas the makers recommend (General Electric X-ray Corporation Direction No. 10976) not more than five steps. However, it is evident that five steps lead to appreciable and

unnecessary discrepancies. It is practicable to use a maximum of two steps.

## (2) DEAN 150-kV SUPERFICIAL SET. HALF-WAVE RECTIFICATION, UNSMOOTHED

A simplified circuit of this set is shown in Fig. 5, from which it will be seen that this is an example of

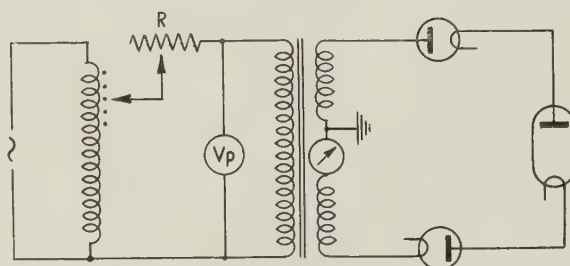


Fig. 5. Dean 150 kV superficial set (simplified circuit)

unsmoothed half-wave rectification. In this case, R is not graduated and the set is intended to be run with R nearly all out. Two sets of readings were taken, (a) with R nearly all out, and (b) with R nearly all in, and the results are given in Table 2. In all cases the kilovoltage was adjusted so that the meter read as nearly 90 kV as possible and the tube current was controlled manually at 4 mA. All measurements were taken with full back-scatter, using a Victoreen.

From Table 2 it will be seen that a resistance in the primary circuit leads to a lower output and the effect is more marked when the radiation is more heavily filtered as in the case of the Maximar.

Cathode-ray oscillograph photographs were taken of the tube current and voltage wave-forms, using the circuit shown in Fig. 6, where  $R_A = 10\text{ K}$ ,  $R_B = 1\text{ M}$ , and  $R_C = 400\text{ M}$ . The resulting wave-forms are shown in Fig. 7(a) and (b), with R nearly all out and Fig. 7 (c) and (d) with R nearly all in. The kilovoltage readings were 90.5 with R nearly all out and 90 with R nearly

Table 1. Maximar 220-kV deep therapy set

Added filtration 1 mm Cu + 1 mm Al +	Resistance control at stud 23			Resistance control at stud 16			Percentage loss in output due to increased R
	Output, r/min	H.V.L., mm Cu	Equiv. kV	Output, r/min	H.V.L., mm Cu	Equiv. kV	
0	31.2	1.70	97	29.4	1.59	94	6
1 mm Cu	20.0	2.34	118	18.6	2.23	114	7
1.5 mm Cu	16.1			14.8			8
2 mm Cu	14.4	2.60	127	13.0	2.50	123	10
3.5 mm Cu	9.4			8.3			12

Table 2. *Dean 150-kV superficial set*

Added filtration	<i>R</i> nearly all out		<i>R</i> nearly all in		Percentage loss in output due to increased <i>R</i>
	Output, r/min	H.V.L. mm Al	Output, r/min	H.V.L. mm Al	
Nil	211	1.58	160	1.35	24
2 mm Al	89		60		32
4 mm Al	54		35.5		34
0.25 mm Cu	30.4		17.2		43
0.5 mm Cu	15.3		7.3		52

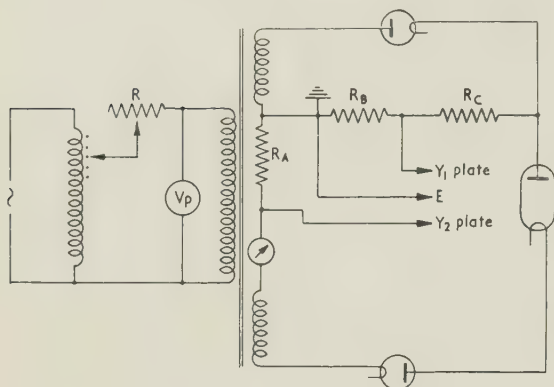


Fig. 6. Dean circuit for obtaining wave-forms

all in. The ripples are presumably due to the capacity of the cables which are several yards in length between rectifier and X-ray tube. Fig. 8 shows the primary voltage wave-forms. It is instructive to compare these wave-forms, which are those across the h.t. transformer, and show the expected lower peak kilovoltage and increased peakiness when *R* is large, with the tube wave-forms. Here the presence of a larger value of *R* still leads to a decrease in peak kilovoltage, but the voltage drop across the rectifiers is such that the tube voltage wave-form does not differ very much from a sine wave; indeed, it differs less than when *R* is small. This is consistent with the theoretical explanation for this type of set. It is apparent from Fig. 7 that the current

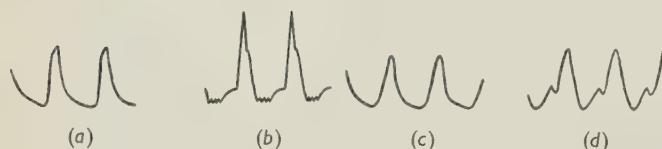


Fig. 7. Dean tube voltage and current wave-forms

- (a) *R* nearly all out. Tube voltage.
- (b) *R* nearly all out. Tube current.
- (c) *R* nearly all in. Tube voltage.
- (d) *R* nearly all in. Tube current.

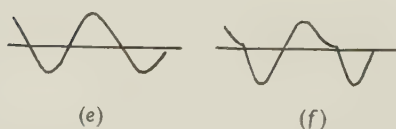


Fig. 8. Dean primary voltage wave-forms

- (e) *R* nearly all out on load.
- (f) *R* nearly all in on load.

changes are large and in this connexion it is interesting to note that if the tube current is correctly adjusted to 4 mA for conditions when *R* is nearly all out and then the filament control left unaltered while the auto-transformer is readjusted to give 90 kV with *R* nearly all in, then the tube current reads 4.5 mA. Conversely, adjusting for 4 mA with *R* nearly all in and then readjusting the auto-transformer and *R* so that the latter is nearly all out, the tube current reads 3.5 mA. Thus it is evident that the filament current has to be adjusted so that the filament runs cooler when *R* is nearly all in, in order that the tube current may be kept at the chosen reading. It also appears from the wave-forms that when *R* is nearly all in, the current flows for a longer part of the cycle than when *R* is nearly all out and this is consistent with the filament temperature changes which are necessary.

### (3) WATSON DIAGNOSTIC SET. FULL-WAVE RECTIFICATION, UNSMOOTHED

Fig. 9 is a simplified diagram of this set. It does not normally have a resistive control, but a 20  $\Omega$  variable resistance was put in the primary circuit for these experiments.

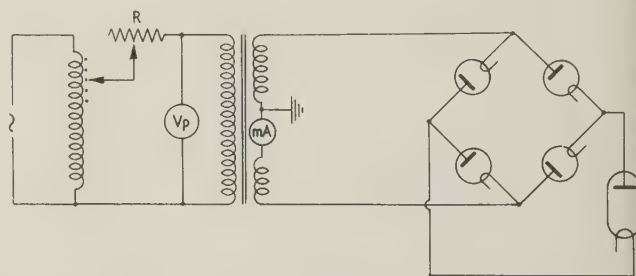


Fig. 9. Watson circuit (simplified) full-wave rectification, as modified

Readings were taken with different values of *R*. In all cases the voltage on load was adjusted to 161 V, using a moving iron voltmeter in the primary circuit and the tube current was manually controlled at 5 mA. The peak kilovoltage was measured by means of a spark gap. All readings were measured on a Victoreen and there was no back-scatter; the F.S.D. was about 11 cm. The results are shown in Table 3. It will be seen that the peak kilovoltage increased slightly and the output also increased as *R* was made larger. This increase is



Table 3. *Watson diagnostic set*

Type of rectification	Volts off load	Peak kilovolts	Approx. $R$ in ohms	Victoreen reading $r/10$ sec
Full wave	161	70.5	0	47.7
Full wave	178	71	6	49.1
Full wave	192	71	12	51.4
Half wave	161	70	0	36.9
Half wave	178	68	5	33.2
Half wave	192	67	10	31.0

presumably due to a peaky voltage wave-form, though this is not obvious from the tube wave-forms which are reproduced in Fig. 10. The circuit used to obtain these is shown in Fig. 11, where  $R_A = 40$  M and  $R_B = 0.25$  M. The oscillograph voltage gain was kept

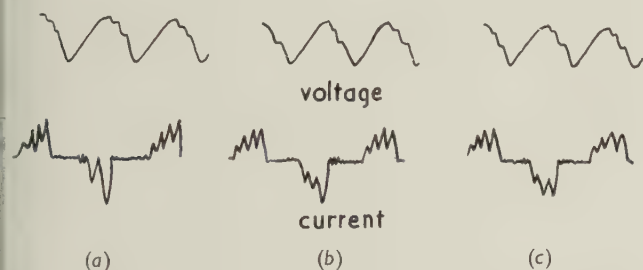


Fig. 10. Watson tube wave-forms. Full-wave rectification  
(a)  $R = 0$ . (b)  $R = 6$  ohms. (c)  $R = 12$  ohms.

constant, but the current gain was altered to avoid limiting. The oscillographs are somewhat difficult to interpret owing to the amount of ripple present. This is due to the large capacities of the cables used; their positions are shown in Fig. 12, where A, B, and C are 8 ft in length and D is 26 ft. As in the case of the Dean set, the current flows for a longer fraction of the cycle as  $R$  increases.

#### (4) WATSON DIAGNOSTIC SET, HALF-WAVE RECTIFICATION, UNSMOOTHED

This is the same set as that described above, with two of the rectifiers removed to convert it to half-wave rectification. It will be noticed that this is now essentially the same circuit as that of the Dean 150-kV set. Conditions were the same as when the set was working with

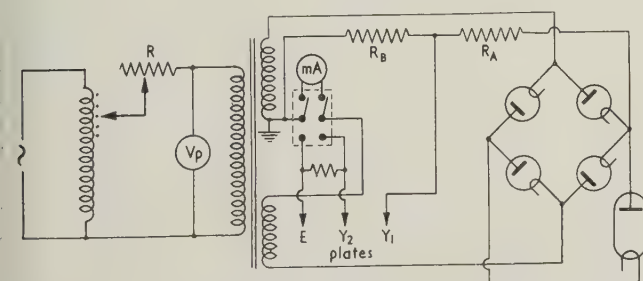


Fig. 11. Watson circuit for obtaining wave-forms, as modified  
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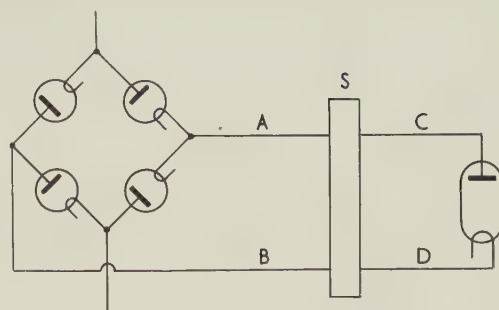


Fig. 12. Cables used with Watson set

full-wave rectification and the results are given in Table 3. Here the peak kilovoltage decreases and so does the output, as  $R$  increases, so that the set now behaves like the other sets with half-wave rectification and unlike this set when working with full-wave rectification.

The cathode-ray oscillographs of tube current and voltage shown in Fig. 13 were taken under conditions as described for the Watson set with full-wave rectification. It is evident that here also the current flows for a longer fraction of the cycle as  $R$  increases.

As the tube current was kept constant at 5 mA for both full-wave and half-wave rectification, it is evident that the filament must have run much hotter in the latter case. This is also clear from the output figures, where the output per conducting half-cycle is proportional to 23.8 ( $R = 0$ ) for full-wave rectification and to 36.9 ( $R = 0$ ) for half-wave rectification.

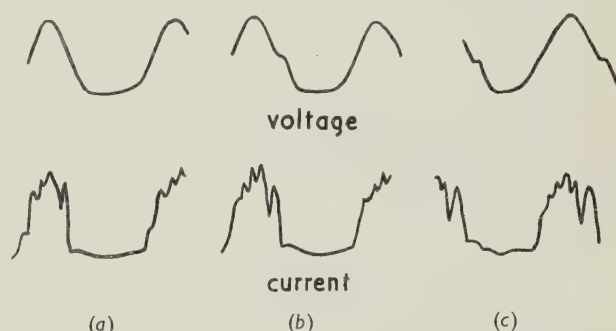


Fig. 13. Watson tube wave-forms. Half-wave rectification  
(a)  $R = 0$ . (b)  $R = 5$  ohms. (c)  $R = 10$  ohms.

#### (5) PHILIPS' METALIX CONTACT SET, 50-KV HALF-WAVE RECTIFICATION, SMOOTHED

Fig. 14 is a simplified circuit of this set, from which it will be seen that there is half-wave rectification with some smoothing. The variable resistance  $R$  was added for the purpose of these experiments.

When this variable resistance was zero, the output was 83.5 r/min., and when the resistance was increased to 14.5  $\Omega$ , the output was 72.8 r/min., a decrease of nearly 13% in output.

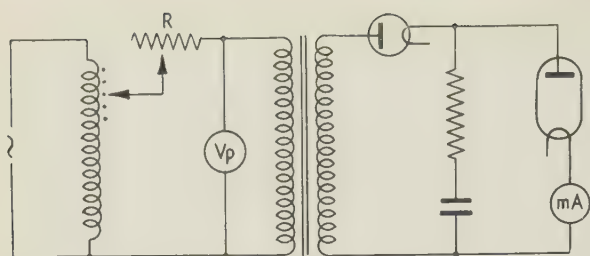


Fig. 14. Philips' metalix contact set as modified (simplified circuit)

Oscillographs of the primary voltage wave-forms are shown in Fig. 15; the set is used in conjunction with a

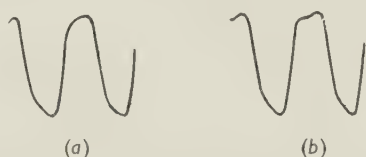


Fig. 15. Metalix primary voltage wave-forms

(a)  $R = 0$  on load.

(b)  $R = 14.5$  ohms on load.

constant voltage transformer, which accounts for some distortion. There is little difference between the wave-forms for  $R = 0$  and  $R = 14.5$  ohms, though the latter is slightly more peaky. The tube voltage and current wave-forms are shown in Fig. 16 and the circuit used to obtain these in Fig. 17. The peak kilovoltage is least when  $R$  is greatest and the current is a little smoother in this case.

This set thus resembles half-wave unsmoothed

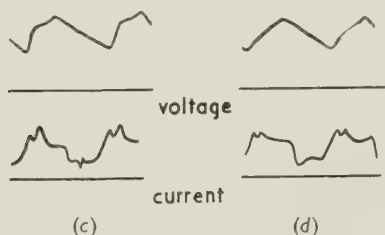


Fig. 16. Metalix tube wave-forms

(c)  $R = 0$ .

(d)  $R = 14.5$  ohms.

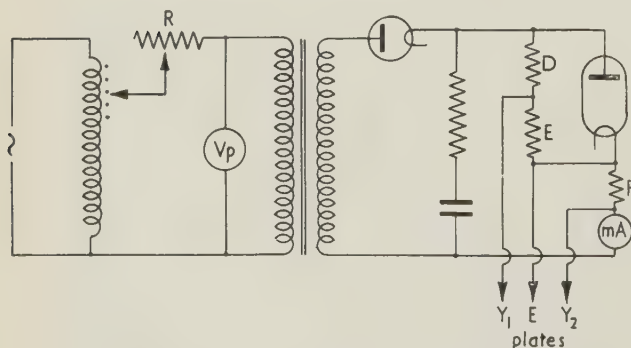


Fig. 17. Metalix circuit for obtaining wave-forms, as modified  
 $D = 400$  M.  $E = 1$  M.  $F = 20$  K.

rectification in behaviour, which is to be expected from considerations of its imperfect smoothing.

## DISCUSSION

Though important changes in output occur when a variable resistance in the primary circuit is used as a fine voltage control, no rules regarding the changes can be given which have universal application. There are two factors to be considered in anticipating the effect. The first of these is the change in "time-average" voltage and this, when present, is of paramount importance. It is, however, present only in sets with self- or half-wave rectification. The second factor is due to changes in wave-form and all types of set show this. At the same time there are, of course, wave-form distortions present due to other causes, such as harmonics introduced by the transformer, and these will be particularly marked in unbalanced circuits as are found in half-wave rectification.

In general, it is likely that self-rectified and half-wave rectified (unsmoothed) sets will show a decrease in output quantity and a softening of the quality, with increased use of resistance, while full-wave rectified sets may show the opposite. The magnitude of the effect, however, cannot be estimated without a detailed knowledge of circuit constants and should always be measured.

It is important that these variations in quality and quantity of output should be eliminated from both physical and clinical work and this can be done by specifying the off-load voltage. In practice it is best to specify this voltage at the lowest practicable value, so that  $R$  is also a minimum when the correct voltmeter reading on load is attained, as this arrangement has the following advantages:

- (1) Voltmeter most nearly pre-reading.
- (2) Least heating of variable resistance.
- (3) Greatest X-ray output (self- and half-wave rectification).
- (4) Hardest X-ray output (self- and half-wave rectification).

This practice has been carried out for over two years with a Maximar 220-kV set with completely satisfactory results.

## ACKNOWLEDGEMENTS

It is a pleasure to acknowledge the following assistance: Mr. E. J. Steadman of Watson and Sons (Electro-Medical) Ltd., for putting an X-ray set at the authors' disposal, modifying it and personally assisting with the measurements; Dr. F. W. Spiers for helpful discussion of the results.

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# The Diffusion of Moisture through Flax Seed

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Measurements of the variation of moisture content through a bulk of flax seed have been made over a period of one year. Each bulk sample of seed had a moisture content initially uniform throughout its volume and the surface was exposed to an atmosphere of 75% relative humidity. The moisture content of the seed was determined gravimetrically. The results indicate that the controlling factor in the movement of water through the seed is diffusion of water vapour through the interstitial air spaces, but owing to the heterogeneous nature of the material, a diffusion coefficient cannot be uniquely defined. An empirical relationship was found to apply for a limited range of conditions.

## 1. INTRODUCTION

During the period 1943-46, an extensive investigation to the storage of various products of the flax processing industry was undertaken at H.M. Norfolk Flax Establishment.<sup>(1-5)</sup> The temperature and moisture relations of flax straw, fibre and seed at the various stages of processing were studied in order to determine the most suitable storage conditions for the retention of quality. The work reported in this paper formed part of the investigation.

Flax seed, immediately after removal from the straw, usually has a moisture regain between 11% and 16%. It is well established<sup>(6)</sup> that seed stored in this condition may deteriorate fairly rapidly, and a regain of 10% is regarded as the limit for safe storage. It is usual to dry the seed in a stream of hot air at the time of cleaning, so that the regain is reduced to between 7% and 9%. The seed is stored in bags each containing 1 cwt until it is required for sowing. The period of storage may be a year or more.

The humidity-regain isotherm for flax seed at 20°C, taken from a Special Report to the Ministry of Supply,<sup>(7)</sup>

is shown in Fig. 1. It will be seen that seed at 8% regain will be in hygroscopic equilibrium with an atmosphere of 45% R.H. The atmospheric humidity in this country is often above this figure, and readings taken in a flax seed store over a period of nine months,<sup>(1)</sup> showed fluctuations between 40% and 80%, the average humidity over the whole period being 65%. It will be seen from Fig. 1 that 65% R.H. corresponds to a regain of 10%, and 80% R.H. to a regain of 13%, therefore it is important to know to what extent the moisture content of seed stored in bulk is affected by these fluctuations in humidity.

The work reported below was undertaken to elucidate this point by studying the diffusion of water vapour through a bulk of seed similar in size to that contained in a standard seed bag. From the results of gravimetric determinations of moisture content at different depths below the exposed surface of the seed, at intervals over a period of one year, an empirical equation for the diffusion was deduced. The cases of adsorption and desorption were treated separately.

The term "regain" as used here denotes the moisture content of the seed expressed as a percentage of the dry weight, under conditions of equilibrium. It is not usual to apply the term in cases where the moisture content is changing. In this paper "moisture content," expressed on a dry weight basis, is used when referring to non-steady conditions and "regain" is used in reference to steady conditions only. It should be pointed out that the convention normally adopted for flax seed is that laid down in the International Rules for Seed Testing, i.e. moisture content expressed as a percentage of the initial weight of the sample, but in the present case this would only complicate the calculations and lead to confusion.

## 2. THEORY

The fundamental law of diffusion of a fluid through a homogeneous medium, Fick's Law, states that the quantity of fluid diffusing through unit area in unit time is proportional to the concentration gradient normal to the area. For the case of diffusion in the  $x$ -direction only, the law may be expressed by the equation

$$\frac{dQ}{dt} = -D \frac{dc}{dx} \quad (1)$$

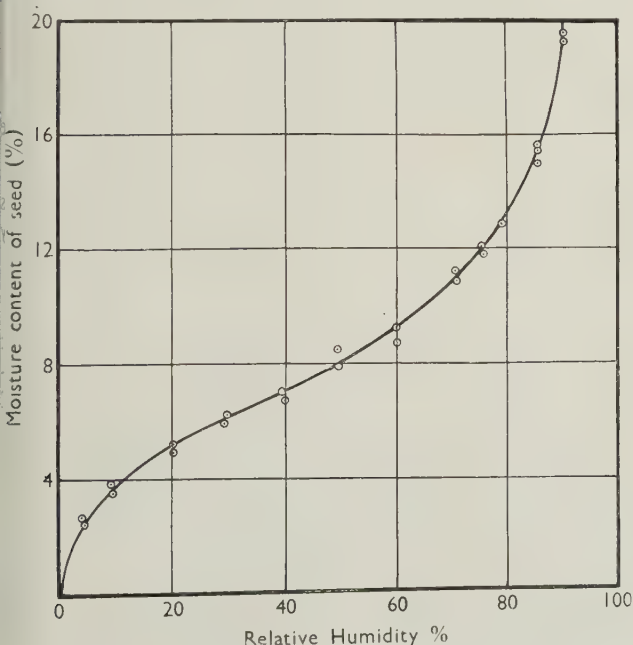


Fig. 1. Equilibrium moisture content of flax seed.

where  $Q$  is the mass of fluid diffusing through unit area in the  $yz$ -plane,  $t$  time,  $dc/dx$  the concentration gradient, and  $D$  the coefficient of diffusion (of dimensions  $L^2 T^{-1}$ ).

By consideration of the mass of fluid flowing into and out of an infinitesimal layer of unit area, we obtain from equation (1)

$$\frac{dc}{dt} = D \frac{d^2c}{dx^2} \quad (2)$$

The integral solution of this equation is in the form of an infinite Fourier series, the exact form depending on the boundary conditions of the problem under consideration.

When water vapour enters a bulk of flax seed, besides diffusing through the interstitial air spaces, it is adsorbed by the seed, and may be transmitted from seed to seed by mutual contact. For this reason Fick's Law will not apply exactly to the movement of moisture through the seed. As a first approximation, however, we may treat the bulk of seed as a homogeneous medium and try to find an empirical relation of the same general form as Fick's Law which will describe the diffusion.

In terms of gravimetric moisture content,  $m$ , equation (2) becomes

$$\frac{dm}{dt} = D \frac{d^2m}{dx^2} \quad (3)$$

As the readings of moisture content were not sufficiently accurate or detailed to determine the second differential coefficient with the required accuracy, equation (3) could not be used to determine the value of  $D$ . In order to use equation (1), it is necessary to evaluate  $Q$ , the quantity of water diffusing through unit area, from the values of moisture content at different levels. In terms of the percentage moisture content, equation (1) becomes

$$\frac{100}{\rho} \frac{dQ}{dt} = -D \frac{dm}{dx} \quad (4)$$

$\rho$  being the bulk density of the seed.

Initially all the seed has a uniform moisture content. Let this be  $m_0$ .  $Q$  is the total mass of water which has passed through unit area in the  $yz$  plane, at a distance  $x$  from the surface of the seed, in an interval  $t$  from the commencement of diffusion, so that if  $l$  is the total depth to which diffusion has taken place

$$Q = \int_x^l \frac{m - m_0}{100} \rho dx$$

or

$$\frac{100Q}{\rho} = \int_x^l (m - m_0) dx \quad (5)$$

Thus equation (5) enables  $Q$  to be evaluated from the experimental results, and equation (4) may be used to find the value of  $D$ .

### 3. EXPERIMENTAL

As was stated in Section 1, flax seed is stored in 1 cwt bags. When filled, these bags are roughly oval in section, the least diameter being approximately 12 in. so that the depth of seed which moisture has to penetrate is about 6 in. In Part I of the investigations the seed was contained in tins, 9 in square by 9 in high, and arrangements were made to take samples of seed from different depths in the tin to a total depth of 6 in.

#### PART 1 OF EXPERIMENT

##### General Arrangement

A forty-gallon galvanized iron tank was used as a humidity chamber, Fig. 2. The temperature of the room in which this was situated was controlled at 20°C. A large photographic dish containing a saturated solution of sodium chloride was placed on the floor of the tank and two tins containing the seed were supported on wooden slats about 4 in above the dish. The top of the tank was covered by a sheet of glass and the edges sealed with plasticine. The sodium chloride solution maintained a relative humidity of 75% in the chamber.

##### Construction of Seed Containers

In order that samples of seed could be removed from the tins without disturbing the main bulk of seed, nine cylindrical containers made of perforated zinc were embedded in each tin of seed. The construction of these containers is shown in Fig. 3. Each was 2 in in diameter and 6½ in long. Zinc having nine perforations to the inch was used. Each container was made up of nine sections, five of these were 1 in high and these were surmounted by two sections ½ in high and two ¼ in high. The bottom section was in the form of a tray

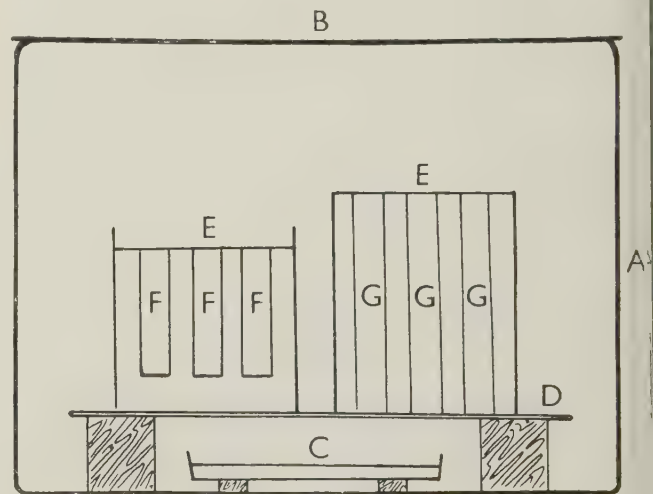


Fig. 2. Humidity chamber

- |                           |   |
|---------------------------|---|
| A 40 gallon tank          | F Zinc containers embedded in seed—Part 1 |
| B Glass cover             | G Zinc containers embedded in seed—Part 2 |
| C Saturated salt solution |   |
| D Wooden slats            |   |
| E Tins containing seed    |   |



and the others were cylindrical rings. The sections were bound together with adhesive cellulose tape,  $\frac{1}{2}$  in wide. The seed was sampled by removing the cellulose tape and sliding the section off sideways, taking the seed with it.

### *Selection of Seed*

The seed used was Liral Prince variety from the 1942 crop. Two quantities of seed weighing 20 lb each were taken from the same flax crop. One was taken before the seed was dried, and had a moisture content of approximately 16%. The other was taken after drying to a moisture content of about 7%. Each lot of seed was thoroughly mixed and kept in an air-tight tin for a week so that the moisture content would be the same throughout the seed. Before filling the containers each lot of seed was again thoroughly mixed. The tins which were to contain the seed were half filled with wet and dry seed respectively, and the perforated zinc containers arranged in the form of a square on the surface. The tins were then filled so that the seed was level with the top of the containers, and finally the containers them-

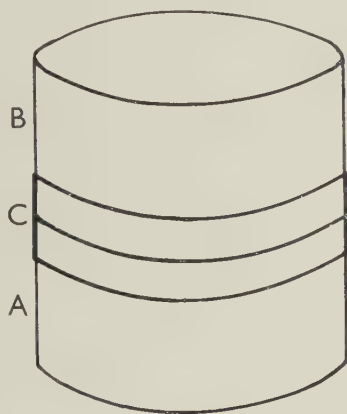


Fig. 3. Construction of seed containers

- A Bottom tray of perforated zinc
- B First ring of perforated zinc
- C Adhesive cellulose tape

selves were filled. In order to find the initial variation of moisture content, one container was removed from each tin directly it had been filled.

### *Method of Sampling*

To remove one of the cylindrical containers without unduly disturbing the rest of the seed, a cylindrical metal tube  $2\frac{1}{4}$  in in diameter and 8 in long was pushed through the seed round the outside of the container, which was then withdrawn. A cylindrical wooden block, the same size as the container was placed inside the tube, and the tube removed. It was possible to withdraw containers for sampling in a very few minutes in this way without removing the tins of seed from the humidity chamber. Having removed the containers from both the wet and the dry seed, samples were taken for moisture content determinations in the following

way. Two samples, each weighing about 2 g, were skimmed from the surface of the seed and placed in stoppered weighing bottles. This removed seed to a depth of about  $\frac{1}{8}$  in. The cellulose tape holding the first  $\frac{1}{4}$  in ring of the container was removed and this section slipped off sideways, carrying the remaining seed with it. The freshly exposed seed was then sampled as before. This procedure was continued until all sections of the containers had been sampled. When samples had been taken from both the wet and dry seed the bottles were weighed and placed in a thermostatically controlled oven at  $103^{\circ}\text{C}$  for five hours. After being removed from the oven the bottles were placed in a desiccator for 30 min to cool before being weighed again.

Containers were removed for sampling after intervals of 3 days and 1, 2, 4, 8, 16, 32 and 52 weeks. The saturated salt solution was replenished as necessary.

### *PART 2 OF EXPERIMENT*

It was found during the course of the experiment, that a temperature gradient of a few degrees existed between the top and bottom of the humidity chamber, and it was thought that this might influence the results. Also, to verify the theoretical conclusions of Section 2, it would be necessary to measure the total mass of water diffusing through the seed, and from equation (5) this requires a knowledge of the moisture content throughout the whole depth of seed. The experiment was therefore repeated with modifications to take account of these considerations.

The general arrangement was the same as in Part 1. The tins containing the seed were 12 in high, the bottoms being removed and both ends covered with sacking similar to that used to make seed bags, which was reinforced by four thin steel rods. As before, nine cylindrical containers were embedded in the main bulk of seed. However, owing to the different construction of the tins and the increased depth of seed, it was found impossible to force the metal tube round the outside of a container in order to withdraw it for sampling. The containers were therefore made of sheet zinc instead of being perforated, and the surrounding seed was used merely to support the containers in a vertical position. The containers were constructed in a similar manner to those used in Part 1, but each consisted of 24 identical cylindrical rings 2 in in diameter and  $\frac{1}{2}$  in high, both ends of the container being open. The sections were bound together, as before, with cellulose tape. Two 2 in lengths of 18 S.W.G. copper wire were soldered to each of the end sections, to anchor the containers to the sacking.

The seed was selected and prepared in the same way as in the first experiment, the moisture content of the wet and dry seed being approximately 14.5% and 6% respectively. When filling the tins with seed, the containers were first fixed in position by pushing the end wires through the bottom sacking and twisting them together. The tins and containers were then filled to

the top with seed and the top cover of sacking placed in position and fastened to the containers by means of the wires. To remove a container for sampling, the top sacking was first lifted off and a metal lid fixed on the end of one container with cellulose tape. The sacking was then replaced and secured again leaving the end wires of the chosen container unfastened, and the whole tin inverted. After removing the bottom sacking, the chosen container was removed. As the level of the surrounding seed fell when a container was removed in this way, more seed was added to bring the level up to the top of the tin. Samples were removed from each of the 24 sections of the container by the method described in Part 1. Samples were taken at the beginning of the experiment and at the same intervals as previously, that is, after 3 days, and 1, 2, 4, 8, 16, 32, and 52 weeks.

#### 4. RESULTS

The values of moisture content obtained in Part 1 are given in Tables 1 and 2, and those obtained in Part 2 in Tables 3 and 4. It can be seen that the results are affected by a drift in the general level in some cases, this is most marked in Part 2, amounting to some 3% for  $t = 1$  week. The readings for 3 days, 1 week, and 2 weeks are involved in Part 2 and those for 1 week in Part 1. A correction was made for this error, which was probably due to a slight change in the technique of moisture content determination.

Fig. 4 shows, for the case of adsorption in Part 2, the change in moisture content with depth at intervals

of time ranging from 3 to 300 days after the beginning of diffusion. These curves were obtained by graphical smoothing of the experimental results in the following manner. The experimental values of moisture content were plotted against time for each level at which a sample was taken, and a smooth curve drawn through each set of points. The curves of Fig. 4 and similar sets for the other three cases were constructed from these. In Part 2 the mean values of the moisture content for pairs of points at equal distances from the two exposed surfaces, were taken. This compensated for the slight temperature gradient mentioned in Section 3.

Values of the quantity  $100Q/\rho$  were derived from these curves by the use of equation (5) of Section 2. The integration was performed graphically by means of a

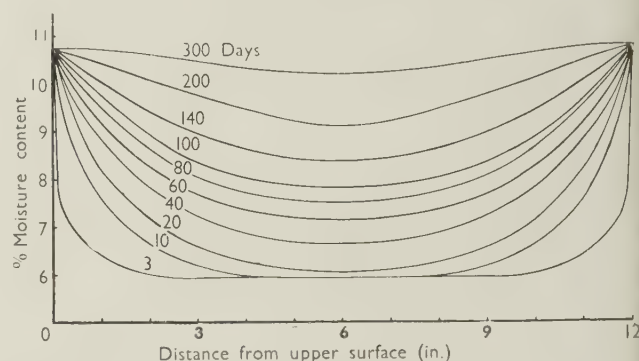


Fig. 4. Distribution of moisture through seed. Part 2—Adsorption

Table 1. Part 1 of Experiment—Desorption. Percentage moisture content of seed

Mean depth in seed (in)	Time from beginning of diffusion								
	0 days	3 days	1 week	2 weeks	4 weeks	8 weeks	16 weeks	32 weeks	52 weeks
$\frac{1}{16}$	15.8	12.0	11.1	11.7	11.8	11.2	11.1	11.4	11.5
$\frac{5}{16}$	15.8	13.3	12.8	11.4	11.8	11.3	11.2	11.5	12.1
$\frac{9}{16}$	16.0	13.9	13.7	12.5	12.1	11.6	11.3	11.7	12.4
$1\frac{1}{16}$	16.2	14.9	14.8	13.3	12.6	12.3	11.5	11.9	12.5
$1\frac{5}{16}$	15.7	15.2	15.5	14.6	13.4	12.5	11.9	12.2	12.4
$2\frac{9}{16}$	15.6	15.8	15.6	15.6	14.7	14.1	12.5	12.7	12.4
$3\frac{1}{16}$	15.9	15.9	15.6	15.8	15.4	15.1	12.8	13.1	12.5
$4\frac{9}{16}$	15.9	15.9	15.7	15.8	15.8	15.6	13.7	13.5	12.8
$5\frac{1}{16}$	15.9	15.9	15.6	15.8	16.1	16.0	14.4	14.2	13.6

Table 2. Part 1 of Experiment—Adsorption. Percentage moisture content of seed

Mean depth in seed (in)	Time from beginning of diffusion								
	0 days	3 days	1 week	2 weeks	4 weeks	8 weeks	16 weeks	32 weeks	52 weeks
$\frac{1}{16}$	7.1	8.8	8.9	9.8	10.1	10.6	10.2	10.5	10.8
$\frac{5}{16}$	7.3	8.4	8.6	9.3	9.9	10.2	10.3	10.3	10.6
$\frac{9}{16}$	7.2	7.9	8.4	8.9	9.7	9.9	9.9	10.3	10.8
$1\frac{1}{16}$	7.1	7.5	7.9	8.3	9.1	9.5	9.6	10.4	10.8
$1\frac{5}{16}$	7.1	7.3	7.5	7.9	8.8	9.0	9.5	10.3	10.4
$2\frac{9}{16}$	7.1	7.0	7.3	7.45	8.0	8.5	8.8	10.1	10.5
$3\frac{1}{16}$	7.2	7.05	7.2	7.2	7.5	8.0	8.3	9.7	10.1
$4\frac{9}{16}$	7.2	7.3	7.3	7.1	7.4	7.5	8.0	9.4	9.9
$5\frac{1}{16}$	7.2	7.1	7.15	7.1	7.3	7.6	7.75	9.2	9.8



Table 3. *Part 2 of Experiment—Desorption. Percentage moisture content of seed*

Distance from top surface of seed (in)	Time from beginning of diffusion								
	0 days	3 days	1 week	2 weeks	4 weeks	8 weeks	16 weeks	32 weeks	52 weeks
$\frac{1}{16}$	14.5	11.6	12.6	11.9	11.2	11.1	10.9	—	11.2
$\frac{2}{16}$	14.3	13.4	12.9	12.9	11.9	11.7	11.4	—	11.2
$1\frac{1}{16}$	14.4	14.6	14.8	13.4	12.6	12.0	11.8	—	11.5
$1\frac{9}{16}$	14.5	14.8	14.5	14.3	13.2	12.2	12.0	12.0	11.7
$2\frac{1}{16}$	14.5	15.0	14.8	14.5	13.5	12.7	12.5	12.2	11.9
$2\frac{9}{16}$	14.5	14.7	15.0	14.6	14.0	12.9	12.6	12.4	11.9
$3\frac{1}{16}$	14.7	14.8	14.9	14.4	14.3	13.8	13.1	12.7	12.0
$3\frac{9}{16}$	14.4	14.8	14.9	14.4	14.5	14.1	13.2	13.0	12.3
$4\frac{1}{16}$	14.4	14.7	15.0	14.7	14.4	14.6	13.5	—	12.4
$4\frac{9}{16}$	14.6	14.6	14.9	14.5	14.5	14.5	13.7	13.2	12.5
$5\frac{1}{16}$	14.7	14.8	14.7	14.6	14.5	14.8	13.8	13.2	12.5
$5\frac{9}{16}$	14.5	14.4	14.7	14.8	14.6	14.7	13.7	13.2	12.8
$6\frac{1}{16}$	14.4	14.5	14.8	14.5	14.5	14.0	14.1	13.1	13.0
$6\frac{9}{16}$	14.5	14.6	14.9	14.8	14.5	14.3	14.1	13.1	—
$7\frac{1}{16}$	14.5	14.7	14.8	14.7	14.5	14.8	14.2	13.2	12.6
$7\frac{9}{16}$	14.7	14.4	15.0	14.7	14.5	14.7	14.1	—	12.5
$8\frac{1}{16}$	14.6	14.8	14.9	14.6	14.5	14.5	14.1	12.6	12.4
$8\frac{9}{16}$	14.6	—	14.9	14.4	14.4	14.3	13.7	12.8	12.6
$9\frac{1}{16}$	14.7	14.9	14.8	14.7	14.3	13.7	13.3	12.6	12.7
$9\frac{9}{16}$	14.6	14.5	—	14.6	13.9	13.4	12.9	12.3	12.2
$10\frac{1}{16}$	14.5	14.7	14.9	14.3	13.8	12.5	12.3	12.5	12.3
$10\frac{9}{16}$	14.6	14.9	14.7	13.5	13.1	12.5	12.0	12.0	12.1
$11\frac{1}{16}$	14.5	14.3	13.8	12.9	12.5	12.2	11.5	11.8	12.1
$11\frac{9}{16}$	14.5	13.5	12.6	12.7	11.9	11.8	11.0	—	11.6

Table 4. *Part 2 of Experiment—Adsorption. Percentage moisture content of seed*

Distance from upper surface of seed (in)	Time from beginning of diffusion								
	0 days	3 days	1 week	2 weeks	4 weeks	8 weeks	16 weeks	32 weeks	52 weeks
$\frac{1}{16}$	5.9	7.2	8.9	9.7	—	9.7	10.3	10.8	10.7
$\frac{2}{16}$	6.0	6.6	8.3	8.8	9.4	9.4	10.0	10.8	10.7
$1\frac{1}{16}$	5.9	6.5	7.5	7.8	8.6	8.5	9.4	10.5	10.8
$1\frac{9}{16}$	5.8	6.2	6.9	7.2	7.8	8.5	9.1	10.2	10.7
$2\frac{1}{16}$	6.0	6.1	6.6	6.7	7.5	8.1	8.8	10.1	10.6
$2\frac{9}{16}$	5.9	6.0	6.4	6.7	7.1	7.9	8.5	9.9	10.7
$3\frac{1}{16}$	5.9	5.9	6.3	6.4	6.7	7.8	8.4	10.1	10.6
$3\frac{9}{16}$	5.8	6.1	6.0	6.2	6.7	7.4	8.2	—	10.6
$4\frac{1}{16}$	5.9	6.2	6.3	6.3	6.4	7.2	8.1	10.2	10.5
$4\frac{9}{16}$	5.9	6.1	6.4	5.9	6.5	7.4	8.1	9.3	10.7
$5\frac{1}{16}$	6.1	6.1	6.4	6.0	6.5	—	7.9	9.2	10.5
$5\frac{9}{16}$	6.0	5.8	6.1	6.1	6.5	7.2	7.8	9.1	10.6
$6\frac{1}{16}$	5.9	6.1	6.1	6.1	5.9	6.9	8.0	9.4	10.4
$6\frac{9}{16}$	5.9	6.0	6.2	6.1	6.3	7.0	—	9.3	10.3
$7\frac{1}{16}$	5.9	5.9	6.2	6.1	6.3	7.1	8.1	9.3	10.6
$7\frac{9}{16}$	5.9	6.1	6.4	6.2	6.3	7.4	7.8	9.5	10.6
$8\frac{1}{16}$	6.0	6.1	6.3	6.2	6.3	7.4	8.3	9.6	10.6
$8\frac{9}{16}$	6.0	6.1	6.4	6.4	6.5	7.3	8.4	9.9	10.7
$9\frac{1}{16}$	5.9	6.0	6.4	6.6	6.8	7.5	8.6	10.1	10.7
$9\frac{9}{16}$	6.0	5.9	6.6	6.6	7.1	7.9	8.7	10.2	10.8
$10\frac{1}{16}$	5.9	6.2	6.7	6.9	7.7	8.4	9.0	10.4	—
$10\frac{9}{16}$	5.9	6.4	7.0	7.5	7.9	8.8	9.2	10.4	—
$11\frac{1}{16}$	5.8	6.7	7.7	8.4	9.1	9.4	9.7	10.7	—
$11\frac{9}{16}$	5.9	7.0	8.3	9.5	—	10.0	10.2	—	—

planimeter. The upper limit of integration,  $l$ , in equation (5) corresponds, in Part 1 to the total depth of seed, and in Part 2 to the distance of the centre from either surface. Families of curves showing  $100Q/\rho$  as a function of time for different values of the depth,  $x$ , were then plotted.

In Part 1 of the investigation the moisture content was measured to a depth of  $5\frac{1}{2}$  in only, whereas the total depth of seed was 9 in. It was not possible, therefore, to evaluate  $100Q/\rho$  after diffusion had penetrated more than  $5\frac{1}{2}$  in into the seed. In the case of the seed adsorbing moisture this occurred at  $t = 20$  days, before enough readings had been taken to make any calculations. For the seed desorbing moisture the calculation can be made up to 80 days.

The slopes of the curves of moisture content and  $100Q/\rho$  at corresponding points give pairs of values of the differential coefficients in equation (4) of Section 2, so that the value of the coefficient of diffusion,  $D$ , can be found. The values of the differential coefficients are shown graphically in Figs. 5 and 6 for the cases of desorption and adsorption respectively. The slopes of the curves were measured by means of a straight edge, and about thirty pairs of values were determined from each set of results.

In Fig. 5, for desorption, the points lie fairly regularly about a straight line which does not pass through the origin. This implies that the coefficient of diffusion

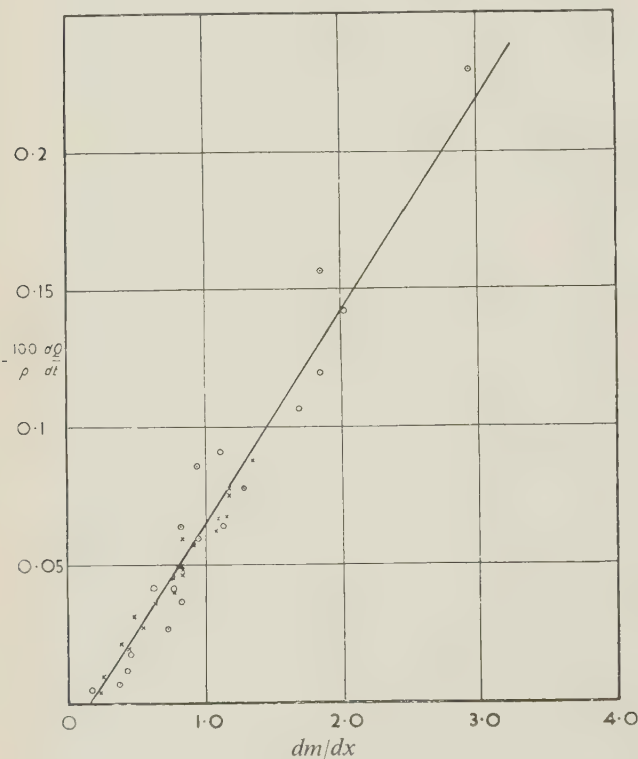


Fig. 5. Values of differential coefficients for the case of desorption

○ — Part 1    △ — Part 2

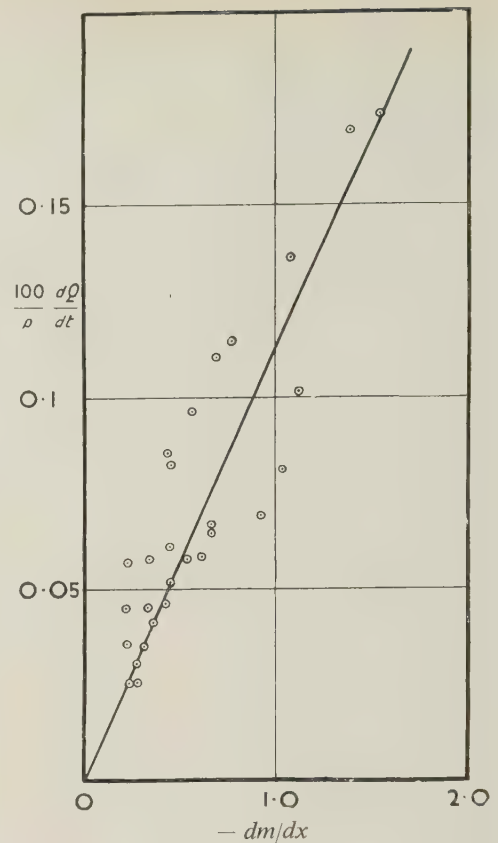


Fig. 6. Values of differential coefficients for the case of adsorption

decreases at very small moisture gradients, and this may be due to the heterogeneous nature of the medium. The modified equation representing the diffusion is

$$\frac{100}{\rho} \frac{dQ}{dt} = A - B \frac{dm}{dx} \quad (6)$$

where  $A$  and  $B$  are constants.

The values of  $A$  and  $B$  are, from Fig. 5,

$$A = 0.012 \pm 0.005 \text{ in/day. } (0.031 \pm 0.013 \text{ cm/day})$$

$$B = 0.078 \pm 0.005 \text{ sq in/day. } (0.50 \pm 0.03 \text{ cm}^2/\text{day})$$

The value of the diffusion coefficient,  $D_d$  as defined by equation (4) varies from 0.023 to 0.073 sq in/day.

For adsorption, Fig. 6, the points are less regular and there is no evidence that the mean line does not pass through the origin. All the points lie above the corresponding line in Fig. 5 indicating that the coefficient of diffusion is greater in this case. The value from Fig. 6 is

$$D_a = 0.114 \pm 0.025 \text{ sq in/day. } (0.74 \pm 0.16 \text{ cm}^2/\text{day}).$$

The limits to the numerical values are rough estimates from the scatter of the individual points in Figs. 5 and 6.

The difference in the values of the diffusion coefficient obtained in the two experiments and the deviation from



ick's Law indicate that the main factor concerned in the movement of water through the seed is the diffusion of vapour through the interstitial air spaces. If this is the case, the rate of diffusion is proportional to the vapour pressure gradient in the air, not to the concentration gradient of moisture in the seed. The relationship between vapour pressure and moisture content of the seed is shown in Fig. 1. For the range of moisture contents covered in the adsorption experiments, 6% to 11%, the graph is fairly linear with an average slope of 0.122% moisture for 1% change in R.H. For the range covered in the desorption experiments, 11% to 20%, the graph is more curved, the slope varying from 0.122% moisture to 0.55% moisture for 1% change in R.H. The diffusion coefficients should be inversely proportional to these slopes, so that the ratio of the coefficients  $D_d/D_a$  should vary between 0.122/0.20 and 0.122/0.55, i.e. between 0.61 and 0.22. The limits of this ratio actually obtained are 0.073/0.114 and 0.023/0.114, i.e. 0.64 and 0.20. This suggests that the exchange of moisture between seed and the immediately surrounding air takes place much more rapidly than diffusion through the interstices between the seeds, a fact which has been confirmed by Gilling,<sup>(9)</sup> who found that single layers of flax seed with an initial moisture content 3% above or below the equilibrium point will equilibrate with the surrounding atmosphere, except at low humidities, within eight to twelve hours. Similar observations have been made by Babbitt<sup>(8)</sup> in the case of wheat.

The difference in the two results would therefore seem to be due to the non-linear relationship between water vapour pressure and seed moisture content, rather than to different mechanisms for adsorption and desorption.

## 5. CONCLUSIONS

The movement of moisture through flax seed is in two parts, diffusion of the vapour through the interstitial air spaces and adsorption by the seed. This makes an exact definition of the diffusion coefficient impossible. An approximate treatment, in which the seed bulk is regarded as quasi-homogeneous, has resulted

in the establishment, for moderate moisture contents (within the range 5% to 11%), of the empirical relationship of equation (6).

## 6. ACKNOWLEDGEMENTS

The authors are grateful to the Chief Scientist, Ministry of Supply, for permission to publish this paper on work carried out at His Majesty's Norfolk Flax Establishment, Board of Trade, whilst the Establishment was still under the direction of the Ministry of Supply.

The investigation was suggested by Dr. R. W. Powell of the National Physical Laboratory to whom, with Mr. D. R. Gilling of this Establishment, the authors are indebted for help and advice.

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# The Pirani Effect in a Thermionic Filament as a Means of Measuring Low Pressures

By L. SPIERS, D.Phil.,\* and W. P. JOLLY, B.Sc.,† University College, Exeter

[Paper received 3 November, 1949]

The Pirani cooling of a tungsten thermionic filament has been investigated down to a pressure of the order of  $10^{-6}$  mm of mercury. The heating voltage applied to the filament to produce a given emission falls as the pressure is reduced and calibration curves of this voltage against the logarithm of pressure suggest that the effect could be used in a simple pressure gauge covering the range  $10^{-2}$  to  $10^{-6}$  mm of mercury and below.

The filament works at such a low temperature that a sudden accidental inrush of air does not affect it, and the change in heater voltage is large enough (several volts over whole range) to be measured by a robust pointer instrument.

A simple ionization gauge consisting of a straight pure tungsten filament of diameter 0.06 mm surrounded by a spiral wire grid and coaxial cylindrical metal anode was exhausted by a single stage mercury diffusion pump. The normal filament operating voltage and current were of the order of 3 V and 1 A respectively. The grid was kept earthed, except during the outgassing of the metal parts of the gauge by electron bombardment, when it was connected to the anode and a 1000 volt supply. The working anode voltage was 165 volts, and the emission current  $90 \mu\text{A}$  measured with a pointer microammeter. The alternating voltage applied to the filament to produce this emission current was varied by means of a Variac transformer.

The pressure in the system was measured with a McLeod gauge, using a cathetometer to set the mercury level accurately and to measure the height of the head of mercury.

## EXPERIMENT

The heating voltage applied to the filament to produce  $90 \mu\text{A}$  emission current with a fixed anode voltage was observed as the pressure in the system was reduced. To produce this emission current it was only necessary to run the filament at red heat, the temperature being estimated to be of the order of  $1000^\circ \text{K}$ .

Three calibrations of the thermionic pressure gauge were carried out: two with air in the system, and the third with hydrogen admitted via a palladium tube.

In the first air calibration the heating voltage was raised to give 2 mA emission from the filament for a few seconds in order to outgas it, immediately before measuring the voltage required to produce the  $90 \mu\text{A}$  emission.

In the second air calibration a different McLeod gauge of bulb volume 500 c.c. replaced that used previously, and a more thorough preliminary out-gassing of the metal parts of the gauge was carried out by strapping the grid to the anode, and drawing 20 mA emission current with 1000 volts applied to the anode. The filament temperature was estimated to be about  $2400^\circ \text{K}$ . During the actual calibration the grid was re-connected to the

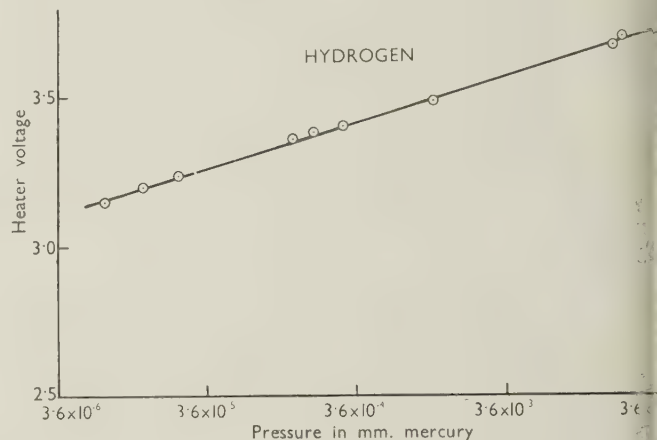


Fig. 1.

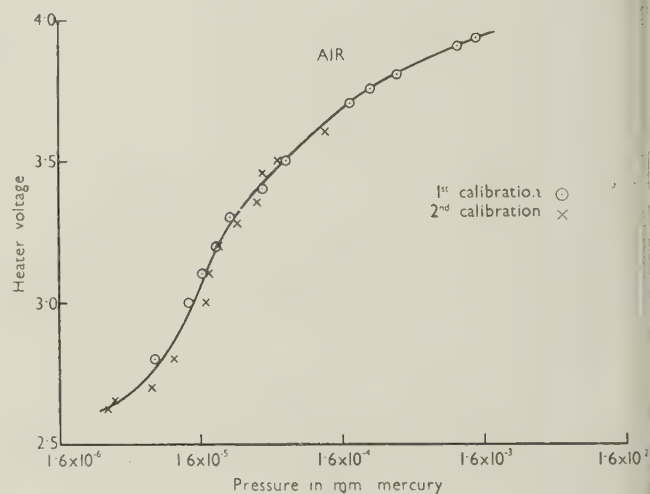


Fig. 2.

filament which was again out-gassed immediately before each reading by drawing 1.5 mA emission for a few seconds, the colour of the filament being pale yellow during this out-gassing.

In the hydrogen calibration the gas absorbed by the filament had a large effect upon the emission obtained.

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† Now at the Royal Naval College, Greenwich.



the filament was accordingly "flashed" at 20 mA emission for a few seconds immediately before taking reading at 90  $\mu$ A emission. This precaution was thought sufficient to ensure a standard clean filament condition for each reading.

The calibration curves obtained are shown in Figs. 1 and 2. In the air calibration agreement between the results from two separate experiments using different McLeod gauges was good. The hydrogen calibration was of special interest since it suggests that a linear calibration may be obtained over a range of pressure as wide as  $10^{-1}$  to  $10^{-6}$  mm of mercury.

A low-pressure gauge employing this effect can cover the range normally needing a Pirani gauge and an ionization gauge. In addition it has the following advantages:—

- (1) Simple construction: a tungsten filament and coaxial anode.
- (2) The two variables observed—heater voltage and emission current—change considerably with pressure, thus allowing robust pointer measuring instruments to replace the expensive and delicate lamp and scale galvanometer.
- (3) The low operating temperature of the filament is a safeguard against damage due to accidental inrushes of air.

This work was originated by one of the authors (L. S.) and carried out jointly; it is surprising that this effect does not appear to have been investigated previously and the work is being continued by one of the authors (W. P. J.) at the Royal Naval College, Greenwich.

## A Method of Measuring the Refractive Index of Liquids

By E. G. KNOWLES, M.Sc., D.I.C., A.Inst.P., and H. JENKINS, B.Sc., A.Inst.P.,\* Nottingham and District Technical College

[Paper received 24 January, 1950]

A method is described for measuring the refractive index of a liquid using only simple apparatus found in any laboratory together with two specimens of known refractive index.

During the course of an investigation, one of the authors (H. J.) had occasion to determine the refractive index of a highly reactive liquid at different concentrations and temperatures. Due to the nature of this liquid, the usual refractometers could not be used since the cement holding the prism in the mount was rapidly attacked and in addition it was necessary to treat the face of the prism with chromic acid to remove any trace of grease and alkali. Recourse, therefore, had to be made to some other method, it being considered that a critical angle method would be most suitable. It was intended, if the method proved satisfactory, to develop the design but preliminary trials were so successful that further development was not considered worth while, the final readings being taken with the initial arrangement of the apparatus. This arrangement, requiring only a reading telescope and prism, is, the authors believe, the virtue of the method.

### ARRANGEMENT OF APPARATUS

The schematic arrangement of the apparatus is shown in Fig. 1, whilst Fig. 2 shows the actual arrangement of the apparatus. The telescope of focal length 16.7 cm with the clamp holding the prism are both fixed to the same retort stand so that relative displacement is avoided; this is essential for reproducible results. The prism is arranged with its lower surface approximately horizontal and the liquid contained in a suitable vessel is brought up so that contact is made between the prism and the liquid surface. The depth of immersion and true

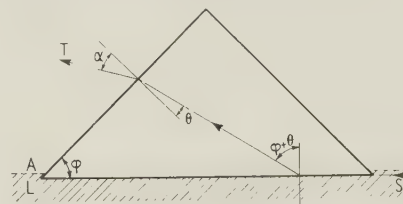


Fig. 1. Schematic diagram of apparatus (A, air; L, liquid; S, from sodium lamp; T, to telescope)

horizontality of the lower surface of the prism are not critical. The telescope is focused for infinity and a sharp boundary is obtained corresponding to the rays refracted at the critical angle. The telescope, previous to mounting, had the "cross-wires" replaced by the glass graticule with a scale of length 1 cm divided into 100 parts.

### THEORETICAL CONSIDERATIONS

We have from the usual law of refraction:

$$N_L = N_g \sin(\phi + \theta) \quad (1)$$

and

$$N_g \sin \theta = \sin \alpha \quad (2)$$

where  $N_L$  and  $N_g$  are the refractive indices of the liquid and material of prism respectively,  $\phi$ ,  $\theta$ , and  $\alpha$  being the angles shown in Fig. 1.

From equation (1) we obtain:

$$\frac{dN_L}{d\theta} = N_g \cos(\phi + \theta)$$

\* Now Extra-mural staff, Leeds University.

and from equation (2)

$$\frac{d\theta}{d\alpha} = \frac{\cos \alpha}{N_g \cos \theta}$$

Combining these

$$\frac{d\alpha}{dN_L} = \frac{\cos \theta}{\cos(\phi + \theta) \cos \alpha} \quad (3)$$

The variation of angle of emergence with change of refractive index of liquid is thus of a complex nature but for small variations of  $N_L$ , which can be determined at any one setting of the prism, it was hoped that a suitable curve could be determined from a few values.

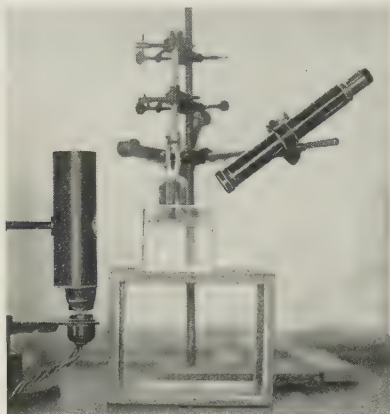


Fig. 2. Apparatus for measuring refractive index of liquids

### RESULTS

A series of 12 mixtures of water and alcohol were prepared, the refractive index of each being determined by an Abbé refractometer to obtain the standard refractive indices. The procedure was to fill the vessel with water, place in position, and adjust the prism until the boundary coincided with the zero of the scale in the eyepiece of the telescope. By means of a pipette a sample was withdrawn and its refractive index determined by the Abbé. The water was then replaced by the first mixture, the reading on the scale noted, and the refractive index determined as before. This process was repeated for the remainder of the mixtures. A prism of refracting angle  $45^\circ$  was used in the first experiments. Results are shown in Table 1 which gives values for the  $D$  line of sodium. The results, on plotting reading against refractive index, give what is, for all purposes, a straight line over this range as is shown in Fig. 3. Using the first and last values of Table 1, the equation to the straight line is

$$N_L = 0.00265R + 1.3333$$

giving the relation between the refractive index of the liquid  $N_L$  and the observed reading  $R$ . This equation was then used to calculate the refractive index from the scale readings with the results shown in the last column. The very close agreement between the calculated and

Table 1. Values for the  $D$  line of sodium (angle of prism  $45^\circ$ )

Mixture	Refractive index $N_L$	Reading ( $R$ )	Refractive index (calculated)
0	1.3333	0	—
1	1.3350	0.60	1.3349
2	1.3366	1.25	1.3366
3	1.3385	1.90	1.3383
4	1.3400	2.50	1.3399
5	1.3420	3.30	1.3421
6	1.3441	4.05	1.3438
7	1.3462	4.80	1.3460
8	1.3485	5.60	1.3482
9	1.3505	6.36	1.3502
10	1.3521	6.95	1.3518
11	1.3539	7.70	1.3537
12	1.3556	8.40	—

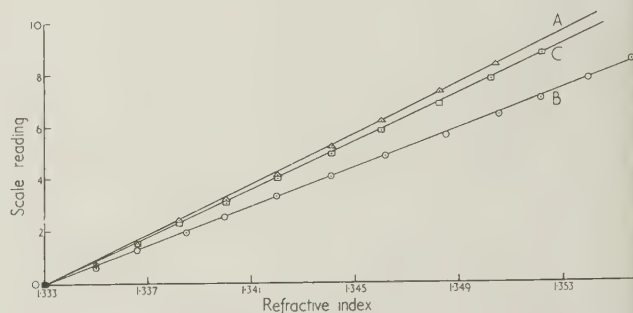


Fig. 3. Dispersion for various angles ( $\phi$ ) of prism.  
A,  $30^\circ$ ; B,  $45^\circ$ ; C,  $90^\circ$

observed values shows that, provided two liquids of known refractive index are available, intermediate values can be obtained with considerable accuracy.

### DISCUSSION

Since  $d\alpha/dN_L$  depends on the angle of the prism, it was thought to be worth while to investigate this. The results for this are shown in Fig. 3, which gives the curves obtained for prisms with angles  $30^\circ$ ,  $45^\circ$ , and  $90^\circ$ . From this it can be seen that if high accuracy is required over a small range, then a prism of  $30^\circ$  angle is chosen, whilst if less accuracy over a greater range is desired, then a prism of  $45^\circ$  is preferable. The fact that the range cannot be extended but that each range should be calibrated separately is shown in Table 2, where the slope  $d\alpha/dN_L$  of the curve is given for different values of  $N$ .

Table 2. Slope of curves for differing values of  $N_L$  (angle of prism  $\phi = 45^\circ$ ;  $N_g = 1.8$ )

Refractive index of liquid	$N_L$	1.30	1.40	1.50	1.60
$d\alpha/dN_L$		1.448	1.612	1.899	2.48

It is thought that the method can be of particular use when refractometers cannot be used or are not available. It also provides a ready method for determining the change of refractive index during a continuous process.



## NOTES AND NEWS

## New Books

**Photoelasticity.** By H. T. JESSOP, M.Sc., F.Inst.P., and F. C. HARRIS, M.Sc., Ph.D. (London: Cleaver-Hume Press Ltd.) Pp. viii + 184. Price 28s. net.

This book combines the function of an introductory textbook with that of a handbook for those engaged in photoelastic work. Several chapters are devoted to the enunciation of fundamental principles without the use of difficult mathematics, some more lengthy proofs being relegated to an appendix. The practical man will find the sections dealing with elementary optics and the specialized optics of the photoelastic bench particularly helpful. Another section gives a clear exposition of the way in which model results can be applied to the structure or engineering part which is presented. The "frozen stress" method of three-dimensional stress analysis and its usefulness are discussed. The book contains a number of examples as well as tables of the mechanical and photoelastic properties of the materials used. A bibliography would have been a useful addition.

A. F. C. BROWN

**Metals Reference Book.** Edited by C. J. Smithells, D.Sc. (London: Butterworth's Scientific Publications.) Pp. xvi + 735. Price 60s. net.

"The object of this *Reference Book* is to provide a convenient summary of data relating to metallography and metal physics. So far as possible the data are presented in the form of tables or diagrams with the minimum of descriptive matter, although short monographs have occasionally been included where information could not otherwise be adequately presented." (Extract from Preface.)

This book covers a wide range of subjects. It begins with a section on First Aid, followed by tables of conversion factors, and then of mathematical formulae. There are then sections on the constants of general and nuclear physics, X-ray crystallography, crystallography, crystal chemistry, geochemistry, metallography, equilibrium diagrams, gas-metal systems, diffusion, general physical properties, thermal, electrical and magnetic properties, mechanical properties of metals and alloys, refractories, fuels, corrosion, welding, brazing and soldering.

There has been a real need for a book summarizing the data of metallurgical science in a form less bulky than that of the A.I.M.M.E. Metals Handbook, and the compression of so much information into a volume weighing only 2½ lb is a triumph of editing and publication for which many readers will be grateful. If such a book could be made accurate and up to date it would be of the greatest value.

In estimating the value of the book in question the reviewer must of necessity be restricted to sections dealing with numerical data with which he is familiar. It must be recognized therefore that such an estimate may be unfair to the book as a whole. The present reviewer is most familiar with the section on Equilibrium Diagrams, and finds that of the 58 diagrams which he knows well, 12 are either in error or badly out of date; this is a high percentage and the section

is undoubtedly weak. The section on ferrous metallography is also out of date. The table of crystal structures contains no mention of the body-centred cubic form of titanium, whilst the lattice spacings of many elements are known to a much higher degree of accuracy than the tables suggest. The section on X-ray crystallography suffers from compression; thus the statement on page 154 suggests that in Debye-Scherrer films the errors resulting from absorption, eccentricity, and faulty calibration of the camera will be removed by extrapolation methods, whereas actually only the first two are so removed. The presentation (p. 140) of wavelengths in kX units, and of density formulae (p. 163) in terms of absolute Angstroms invites confusion. On page 46 the values for the calcite grating space involve one error, whilst another is given with so little definitions that many readers may be puzzled.

It seems clear, therefore, that although invaluable as a rough guide to the data of metallurgical science, the book has not yet reached the consistent accuracy which would entitle it to be considered as a standard work of reference whose values might be quoted in published papers. From the scientific point of view one of the greatest weaknesses of the book is that in so many cases (the chapter on diffusion is an exception) the reader cannot trace the source of the values given. The reviewer is well aware of the difficulties involved, and of the increased size required if references were included. He feels, however, that for most scientific work the investigator must know the source of the figures he uses, not only as an indication of their reliability but also to know whether more recent values are available. For the scientist the most satisfactory solution would probably be to split up the volume into several parts, each of reasonable size and containing some system of reference to original sources. If, however, the opposite view is taken it is clear that more careful searching of the literature is required in several sections of the book in its present form. At the same time it must be acknowledged that this is an ambitious project and many will be grateful to the authors for the work they have done.

W. HUME-ROTHERY

**The Science of Weather.** By D. CHILTON, B.Sc., A.Inst.P. (London: His Majesty's Stationery Office.) Pp. 32. Price 1s. net.

This excellent brochure is the handbook for an exhibition held at the Science Museum in London from March-June 1950 to mark the hundredth birthday of the Royal Meteorological Society. Besides being a handbook for the exhibition it is in itself a useful illustrated brief outline of meteorology. The chapter on "Observing the Weather" deals with surface and upper air observation and special observations such as atmospheric and marine and aircraft observations. A brief chapter on forecasting is followed by one on "Weather in our Daily Lives." The last chapter deals with research in this country under the following headings: cloud physics, turbulence, radiation, upper atmosphere, atmospheric sounding, synoptic meteorology.

**Electrical Measurements and the Calculation of the Errors Involved.** Part I. By D. KARO. (London: Macdonald and Co., Ltd.) P.p viii + 191. Price 18s. net.

The author's intention is to describe the usual d.c. measurements and to assess the value of each by calculating the errors involved. The importance of careful estimation of errors should certainly be impressed on students, but a pre-occupation with this has spoilt the present book. When a quantity  $F$  is determined from measurements of other quantities  $X, Y, \dots$ , the limiting error  $f$  of  $F$  can be calculated in terms of the errors  $x, y, \dots$  of  $X, Y, \dots$ . This calculation is repeated in wearisome detail for each method described to the exclusion of vital practical details as, to give a single example, the selection of the best galvanometer for an experiment from the dozen or so in any manufacturer's catalogue. Electrostatic and magnetic measurements are omitted, but some less familiar uses of the fluxmeter and differential galvanometer are given.

T. B. RYMER

**Automatic Control** (Stetige Regelvorgänge). By WINFRIED OPPELT. (Hanover: Wissenschaftliche Verlagsgesellschaft jointly with Wolfenbüttel: Wolfenbütteler Verlagsgesellschaft G.m.b.H.) Pp. 144.

The present volume, though intended by the author as an introduction into the field of automatic control, can perhaps justly be regarded as a handbook on its practical aspects. The book is set out in a pleasing, systematic manner, and a considerable amount of information is imparted within its hundred and forty pages.

A brief survey of the mathematical foundations is given in the introductory chapter and the use of the Nyquist diagram of the performance of control systems is explained. The reader desiring to make a closer study of the theoretical side of automatic control is referred to the author's companion volume "Principles of Control" ("Grundgesetze der Regelung"), 1947.

Over a hundred pages are devoted to simple control systems. Most of the remaining space is allotted to complex control, and to the construction of scaled-down experimental model-systems. In addition a brief chapter on the scope and classification of manual control provides a stimulating perspective on one's own sensory limitations.

Wherever possible, mathematical relations are illustrated graphically. In fact more than half of the book is taken up by sets of diagrams and graphs, each set appertaining to the analysis of the performance of either a given type of controller or controlled system or of a mechanism consisting of a particular combination of each. Each analysis generally comprises the Nyquist and response diagrams of the system and its units; the (linear) differential equations of the dynamics of the controller and the controlled system; a sketch of a practical embodiment of the mechanism, i.e. for liquid-level control, heat control in furnaces, regulation of power generators, etc., and a verbal discussion relating to its construction and mode of operation.

The determination of the equation of the Nyquist diagram from experimental data is explained, which, together with several brief but illuminating examples, dispersed throughout the text, confirms the author's endeavour to emphasize the concrete rather than the abstract. In this he has certainly

succeeded, and technical readers of the book will no doubt appreciate the effort.

P. FELTHAM

**Electricity and Magnetism—Classical and Modern.** By G. B. DEODHAR, Ph.D., F.Inst.P., and K. S. SINGWI. (Allahabad: Students' Friends.) Pp. viii + 507. Price Rs. 15.

The authors state that the needs and mathematical equipment of B.Sc. students determined the scope of this introductory account and the method of treatment. The first twenty chapters cover very thoroughly the work usually required for a pass degree and should provide also a sound foundation for honours students; the second half of the book gives a concise outline of the development of modern physics. The standard of writing is good and students will appreciate the well-ordered presentation of the mathematical theory and the worked examples in the text. Dr. Deodhar is Reader, and Mr. Singwi Lecturer, in the Department of Physics at the University of Allahabad.

G. R. NOAKES

## Journal of Scientific Instruments

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## ORIGINAL CONTRIBUTIONS

## Methods of Storing and Handling Liquefied Gases\*

By A. J. CROFT, M.A., and G. O. JONES,† M.A., B.Sc., Ph.D., Clarendon Laboratory, University of Oxford

[Paper received 14 February, 1950]

Pieces of laboratory equipment used in handling liquefied gases are described and factors influencing design discussed, with particular reference to liquefied air or oxygen, hydrogen and helium. Among the items described are storage vessels, vacuum pipe-lines, flow-control valves, level indicators and automatic level controllers.

A method of obtaining liquid air by "conversion" from liquid oxygen is briefly described, and a general account given of the low temperature installation at the Clarendon Laboratory, Oxford.

Supplies of liquid oxygen, nitrogen or air are essential needs in nearly every modern research laboratory, and in a laboratory such as the Clarendon Laboratory at Oxford, which includes a large department devoted to low temperature physics, liquid hydrogen and liquid helium are also used; a more detailed discussion of these needs has been given elsewhere.<sup>(1)</sup> Apart from the fact that the most effective methods for the production of "cold" employ gases as working substance, these liquefied gases may be said to contain a large amount of "latent heat" (as latent heat of evaporation) in very compact form. And since they will remain at constant temperature as long as the pressure above the liquid is kept constant—at their normal boiling-points if stored in open vessels, for instance—it is a relatively easy matter to carry out low temperature experiments which require sources of constant temperature. Of course it is just because these liquids are, in normal use, at their boiling-points, and are colder than their surroundings, that special problems arise in their storage and handling. Excessive inflow of heat is uneconomic, causing too much evaporation and loss of liquid, and in any case the smooth handling of a violently bubbling liquid is a very difficult matter.

In a research laboratory we are not, however, primarily concerned with the economic problem, and it is not proposed in this paper to present detailed figures of performance for the various pieces of equipment to be described. Our main needs are for convenience and reliability in meeting the demands, sometimes irregular and unpredictable, of a large number of research workers for one or other of these liquids, and in order to satisfy these needs much new equipment has been designed and built in this laboratory, and is now in use. A general idea of the whole installation may be obtained from Fig. 1. Some of the individual items, such as the storage vessels, pipe-lines, level indicators and controllers, are described below, but not the liquefiers themselves, many of which have been described elsewhere.<sup>(1,2,3,4)</sup>

The technical problems met with are similar for liquid

air, hydrogen, or helium. Most of the pieces of equipment described here were, however, actually designed for use with liquid air or liquid hydrogen because, as indicated in Fig. 1, liquid helium has usually been used where

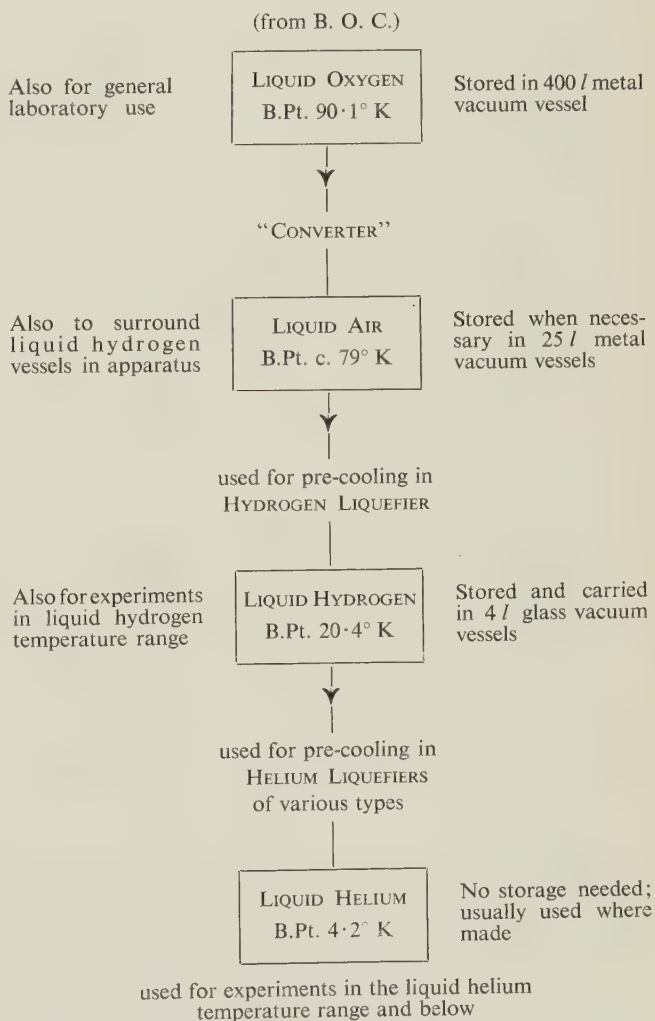


Fig. 1. General plan of the low-temperature installation

\* Based on a paper read before the Low Temperature Group of the Physical Society on 18th January, 1950.

† Now at Queen Mary College, University of London.

made; that is, each piece of apparatus built for experiments at liquid helium temperatures has included its own small helium liquefier. Departures from this procedure are now becoming more common, and a number of new problems are arising.

#### 400 l STORAGE VESSEL FOR LIQUID OXYGEN

Since liquid oxygen had become a commercial commodity and could be obtained regularly and fairly cheaply at the time when low temperature work was begun in this laboratory, it was decided not to build a liquid air plant, but to rely on commercial supplies and to build a large storage vessel for liquid oxygen. This vessel holds about 400 l, and with three weekly deliveries it is always possible to provide liquid oxygen "on tap," in spite of an average weekly consumption of about 800 l.

The vessel (Fig. 2) was designed and assembled in the laboratory in 1938 by Mr. T. C. Keeley and Prof. F. E. Simon, with the assistance of Dr. K. Bosch. It consists of a cylindrical vessel of welded copper suspended in an evacuated space from cables which are linked by means of vitreous insulators, and completely surrounded by four radiation shields, also of copper. The high vacuum is contained in an outer copper cylinder, and the vacuum or "Dewar" vessel thus formed is enclosed in a converted railway engine boiler (kindly presented by the Great Western Railway Company) in which is maintained a "rough" vacuum of a few millimetres of mercury, and which thus supports nearly all the stress due to the external atmospheric pressure. The danger of damage due to unequal contractions has been eliminated by the introduction of a short length of metal bellows tubing in the neck of the inner vessel.

The high vacuum is kept at about  $2 \times 10^{-6}$  mm mercury by means of a continuously running Metrovack Type O3B oil diffusion pump by Metropolitan-Vickers Electrical Co. Ltd., discharging into a 120 l reservoir. Whenever the backing pressure in this reservoir rises to about 0.1 mm of mercury, a rotary pump is automatically brought into action for sufficient time to lower the pressure again to a suitable value.

The "supporting" vacuum in the engine boiler is maintained by the occasional use of another rotary pump. Since this is necessary only about once in every six months, no automatic device is installed. An oil-manometer is, however, connected so as to show the difference in pressure across the outer copper vessel, and this also acts as a safety valve since it blows over if the pressure difference exceeds about 20 cm of oil.

Under these conditions the total loss by evaporation is about 10 l of liquid oxygen per day. The rate of loss actually seems to have increased since the vessel was first assembled, possibly because of condensation of oil vapour on the reflecting surfaces, resulting in increased absorption in the infra-red. We should perhaps emphasize that the quality of the surfaces and the number of radiation shields are important factors in determining

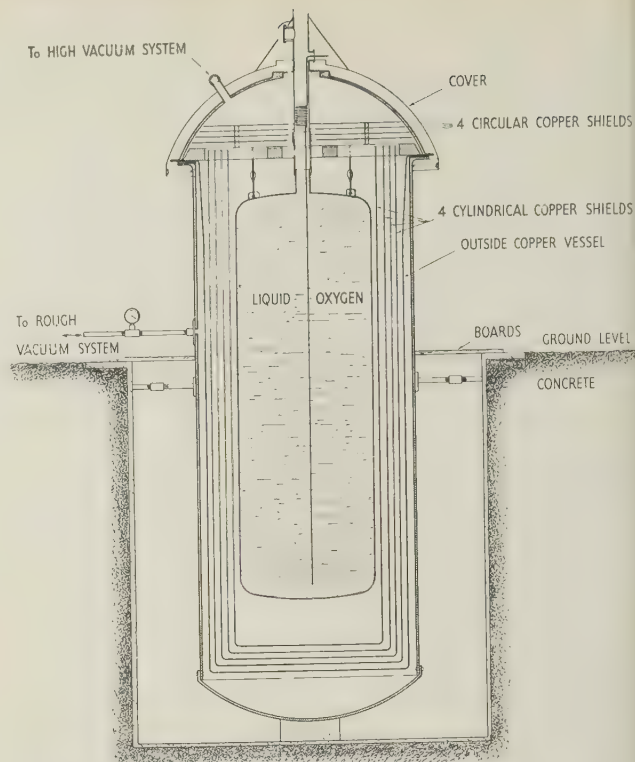


Fig. 2. 400 l vacuum storage vessel for liquid oxygen

the performance; indeed it may easily be shown that if the inner vessel were allowed to absorb black-body radiation from room temperature it would lose all its contents in about 24 h.

At the present time it is necessary to lower siphons into the vessel in order to fill it, or to draw off liquid oxygen, but a permanent "head" is now being constructed which will make this unnecessary and will lead through vacuum siphons to a central supply panel, from which it will be possible to draw off liquid oxygen or (see below) liquid air.

#### OTHER STORAGE VESSELS

About 100 l of liquid hydrogen are used each week, made usually in two "runs" of the hydrogen liquefier carried out on Wednesdays and Fridays. This is served to the individual research workers in 4 l spherical silvered glass vacuum vessels which are made in the laboratory. Since losses by evaporation are rather heavy in these vessels, even when they are surrounded by solid carbon dioxide (about one-third of the contents of a vessel in 24 h), experiments employing liquid hydrogen have usually to be started as soon as it is available. In order to provide liquid hydrogen always "on tap," a large (100 l) metal vacuum storage vessel is now being constructed. This will be more or less similar in design to the larger vessel for liquid oxygen, but in this case the outer wall of the vacuum vessel is to be



re-cooled with liquid air. A point of interest which arises in handling liquid hydrogen is that the "keeping" qualities of the liquid depend on the extent to which conversion to the stable "para-hydrogen" has proceeded. A fuller account of the ortho-para conversion in hydrogen, and of its effect on the rate of evaporation, has been given elsewhere.<sup>(1, 5)</sup>

Other storage vessels used include commercially supplied 25 l metal vacuum vessels for liquid oxygen, air and nitrogen. For use with helium liquefiers and for experiments of all kinds at hydrogen and helium temperatures, silvered glass vacuum vessels of many types are made in the laboratory, often with narrow vertical slits in the silvering which make the contents visible. Containers for liquid helium are made of Monax glasses rather than Pyrex brand glasses, which have too high permeability to helium.

### LIQUID AIR "CONVERTER"

For two main reasons, liquid oxygen is unsuitable for re-cooling in the hydrogen liquefier, and liquid air is used. There is, first, the danger of explosive oxygen-hydrogen mixtures forming in the plant in the event of leaks in the hydrogen circuit. Secondly, the efficiency of the Linde liquefaction process with hydrogen is rather low at temperatures attainable with pumps of practicable size by pumping down liquid oxygen to reduced pressure, though reasonably high at the temperatures attainable with liquid air or nitrogen. (Pure liquid nitrogen is inconvenient because its triple-point,  $63^\circ\text{K}$ , is in the working range.) Liquid air has the added virtue that it boils less violently than liquid oxygen or nitrogen.

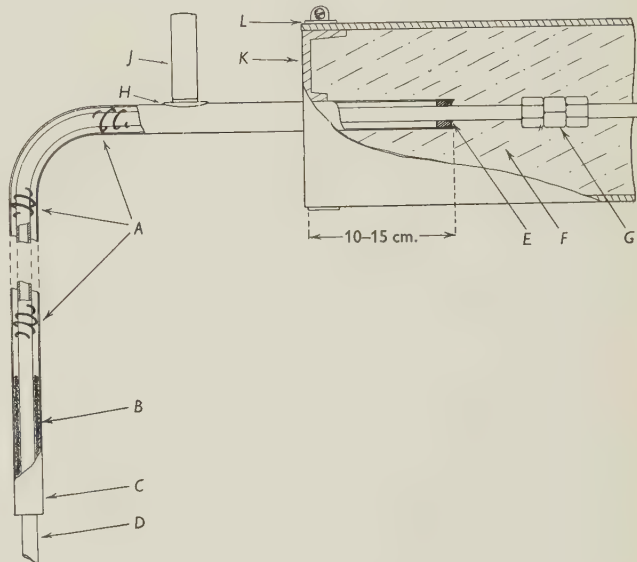
Liquid air is therefore made in the laboratory by condensing compressed air in liquid oxygen. The apparatus in which this is done, usually referred to as a "converter," in effect simply exchanges latent heats between liquid oxygen and liquid air. It consists essentially of a counter-current heat-exchanger and a coil of tubing immersed in liquid oxygen. Air at 10 atm pressure is passed first through a drying train and is then cooled, first in the heat-exchanger by the returning evaporated oxygen and finally in the coil. The liquefied air passes out through a metal siphon (see below) and then through a throttling valve into a container.

The converter in use until recently was designed to be inserted into the liquid oxygen vessel whenever liquid air was required. Severe restrictions were thus imposed on the design of the heat-exchanger, and the necessity for constant handling has been an inconvenience. A new converter is at present under construction. It is contained in its own glass Dewar vessel, draws liquid oxygen from the distribution panel already mentioned, and also serves this panel with liquid air for distribution to the hydrogen liquefier or to small vessels. In this way a heat-exchanger of generous design, and therefore of high efficiency, can be employed. A fuller account is to be published shortly.

An important point to remember in using liquid air is that it enriches itself continuously in oxygen owing to the greater rate of evaporation of nitrogen. Indicators of composition have been found necessary and we have used vapour-pressure thermometers, filled initially with nitrogen or air at about 2 atm pressure, and also hydrometers, for this purpose.

### VACUUM SIPHONS

Double-walled vacuum pipe-lines (usually referred to as "siphons") are used extensively to carry the low boiling-point liquids. These may be of metal or glass, though metal siphons are always used in permanent plant on account of their much greater strength. The hydrogen liquefier, for example, which has three separate stages, is fed with liquid air through one double-walled manifold with three feed-tubes leading to three shorter siphons, one of which enters each stage.



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Fig. 3. Double-walled metal vacuum siphon

The essential features of the design of these metal siphons are illustrated in Fig. 3. The tubes are kept apart by coiled spacers *A* located at intervals where needed. The space between the outer and inner tubes *C* and *D* is evacuated through a tube carried by a mount *H* which is soldered to the outer tube. If there is a high-vacuum pumping system near the point at which the siphon is to be fitted, it may be more convenient to pump this vacuum space continuously, but if not it can be permanently evacuated. In this case a charge of charcoal *B* is used at a portion of the tube which is cold during use, and the side-tube is sealed off and protected by a cap *J* as shown in Fig. 3. The main requirements for successful construction of these permanently evacuated metal siphons are very careful soldering in

making the joints (as at *E*), materials of high quality, and very thorough degassing of the metal surfaces and charcoal before sealing off. Fuller details of the design, and particularly of the method of assembly, have been given elsewhere.<sup>(6)</sup>

The method by which two or more siphons may be connected is also illustrated in Fig. 3. The inner tube of each siphon is fitted with a "petrol" union, and connexion made in an enclosure *F* filled with glass-wool, formed in this case by a rubber tube *L* and a brass collar *K* mounted on each outer tube some distance from the end of the siphon. It is advisable whenever possible to fit a "breathing" tube filled with a drying agent to prevent water from the atmosphere condensing in the glass-wool enclosure.

It is sometimes convenient to use more complicated siphons which include permanent double-walled junctions, since these can be much more compact than junctions of the type illustrated above. They are, of course, more difficult to make; the number of soldered joints must always be kept as low as possible, and special attention paid to their design. The design of the manifold tube mentioned above, which incorporates a 4-way double-walled junction, illustrates some of the most important points, and will be briefly described. The junction is illustrated in Fig. 4. The inner junction was made by passing one tube *A* through two holes drilled in a slightly larger tube *B*, and hard-soldering the two "ring" joints thus formed. The smaller tube already carried two holes, and, as assembled, the centres of these holes lie on the axis of the larger tube so that 4-way communication is established.

The outer wall of the junction was made from a block of brass *C* (shown in section in Fig. 4) carrying two holes drilled at right-angles, into which the outer tubes (*D*, *E*, *F*, *G*) of the four individual branches were hard-soldered. (It was, of course, necessary to carry out the soldering of the inner tubes inside the space formed by these two holes, and before fitting the four outer tubes.) In this case the mount for the sealing-off tube (not

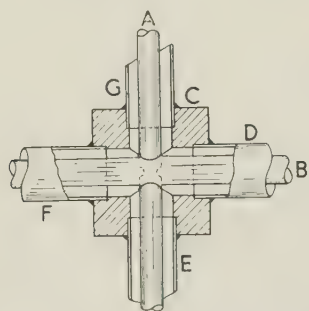


Fig. 4. Four-way double-walled junction for vacuum siphon

shown) was actually turned out of the solid brass in order to save one soldered joint. This particular siphon, which is permanently evacuated, has been used regularly

for over three years without any deterioration in its performance, the outer tubes falling only a few degrees below room temperature when liquid air is passed through the inner tubes.

The choice of material for a metal siphon depends to a large extent on the purpose for which it is to be used. If it is to carry liquid hydrogen, for instance, the tubes must usually be thin-walled (say 0.5 mm or less) and made of some material of low thermal conductivity such as cupro-nickel or stainless steel. For liquid helium, even thinner tubing may be necessary to reduce the thermal conduction and thermal capacity sufficiently, and difficulties are now likely to arise in making satisfactory bends. Fig. 5 illustrates one method by which this

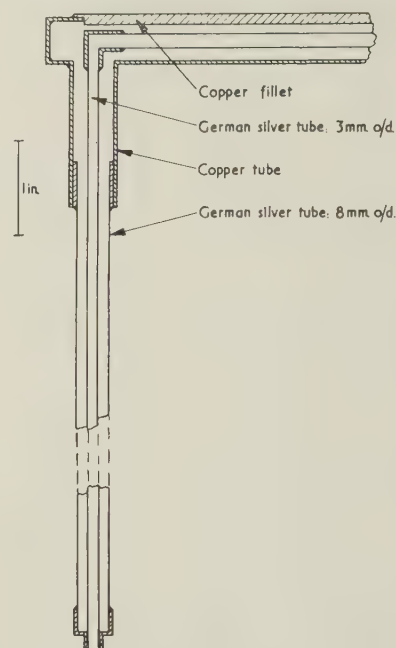


Fig. 5. Metal vacuum siphon for liquid helium

difficulty has been overcome in a siphon for liquid helium. The inner tubing is of German silver of 3 mm diameter and 0.1 mm wall thickness, and the two right-angled bends required (one of which is shown) are built up by the use of small elbows into which the thin-walled tubing is soldered. For assembly, the completed inner tube was inserted into the outer tube through a long slot, which was subsequently closed by soldering in position a metal strip machined to fit the slot. (No spacers are used.) This method of assembly has a number of advantages over the more obvious method of building up the inner and outer tubes in position joint by joint; for instance, it makes it possible to submit the whole inner tube to a rigorous vacuum test before it is fitted into the outer tube. Further details of this siphon and of its performance will be published later.

Small portable siphons, which are not to be used continuously, but only for filling or emptying small



vessels containing liquid oxygen or air, are sometimes made simply by wrapping glass wool around a single tube. Liquid hydrogen is usually transferred from one vessel to another through permanently evacuated double-walled glass siphons made in the laboratory. Although the manipulation of glass siphons requires extreme care, they are very light in weight, do not require re-charge of charcoal, and have the advantage that the liquid hydrogen flowing through the inner tube can be seen. It is proposed to use siphons of similar type for liquid helium, but in this case it will probably be necessary to silver the inner surfaces in order to reduce losses due to radiation.

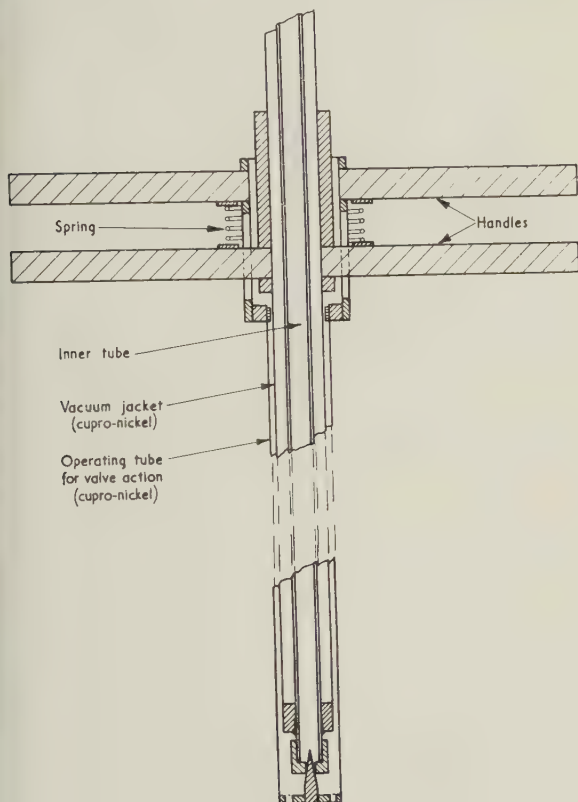


Fig. 6. Valve fitted to a metal siphon

#### VALVES

In order to control the flow of low boiling-point liquids in the plant, all-metal valves are used which are adapted from bellows-sealed valves of standard type as used in high-vacuum work. The main modification is that the moving spindle and outer (fixed) barrel are considerably lengthened by extensions made of some material of low thermal conductivity, so that the base of the valve, through which the liquid passes, may be surrounded by an adequate thickness of glass wool, and the operating handle or knob brought out to room temperature.

It is often convenient to fit a metal siphon with a valve at one of its ends. Such a valve is illustrated in

Fig. 6. A third tube, co-axial with and surrounding the two tubes of the siphon, is movable and carries at its lower end the valve needle. The valve is operated by some device fitted at the upper end of this tube, which is at room temperature. In the valve illustrated in Fig. 6, operation is by means of two handles. Alternatively, a knurled knob and screw thread may be used to transmit the axial motion to the valve needle. It is usually advisable to design the valve in such a way that the closing force is applied by a spring (as in the valve illustrated), and the operating mechanism acts against this spring to open the valve. This prevents damage to thin-walled tubing which might be caused by the application of excessive force when closing the valve, and also prevents erratic operation due to unequal contractions of the tubes. It may sometimes be necessary to fit a bellows seal between the outer tube of the siphon and the operating tube, in order to prevent escape of gas or liquid except through the holes at the bottom of the operating tube.

#### LEVEL INDICATORS

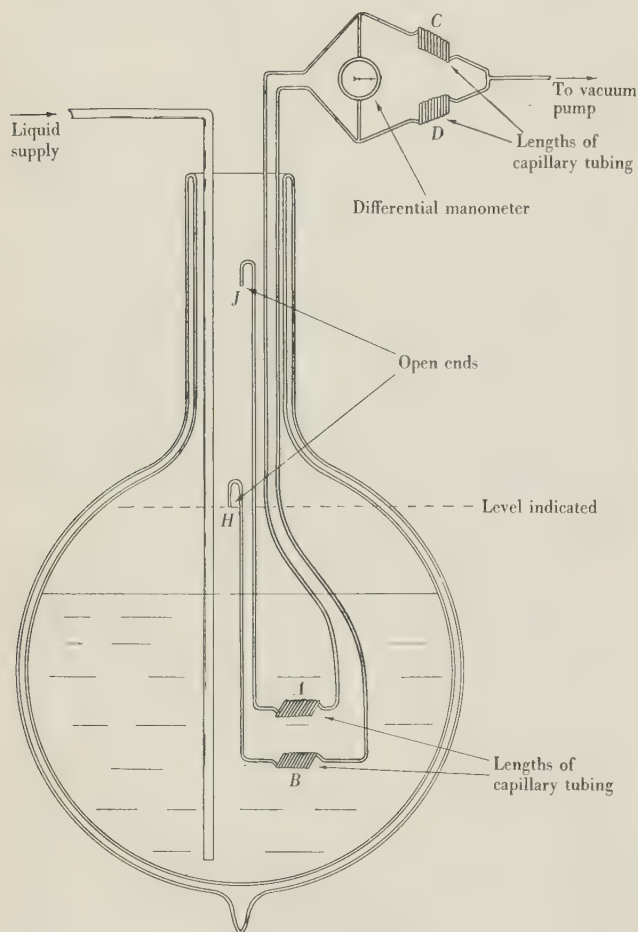
A float made of a length of thin-walled German silver tubing can be used to indicate the level of liquid air in a vessel, and if it is provided with a long neck of smaller diameter it will usually be possible to make the top of the float visible even if the level of liquid is some distance below the top of the vessel. The authors have used floats of this kind, each moving up and down loosely in its own chamber formed by a tube of stainless steel pierced with a number of holes. If the vessel is completely enclosed—if, for instance, it contains liquid air boiling under reduced pressure—this tube may be brought out at the top and terminate in a short length of glass tubing through which the top of the float can be seen. It is of course essential that hollow floats to be used in this way should be completely air-tight, not only from the point of view of accuracy in indication, but also because there is a danger of a minor explosion if a hollow float into which liquid has leaked is allowed to warm up too quickly. It is sometimes useful to attach a small piece of magnetic material to the top of the float so that it may be "tickled" by means of a magnet if it sticks in its tube—as may sometimes happen if ice or other solid enters the system.

The "Hampson-meter," which indicates the level of liquid in terms of the difference in pressure between a point in the vapour above the liquid and a point near the bottom of the vessel, is another convenient form of level indicator. The level of liquid oxygen in the 400 l storage vessel is indicated in this way, and the tube extending to the bottom of the vessel may be seen in Fig. 2. This method has, of course, the disadvantage that, as normally used, it is not direct-reading, but only indicates when the tube extending to the lower level is cleared of liquid by some means such as the application of heat or of the required over-pressure of air.

Both the above methods of indicating liquid levels

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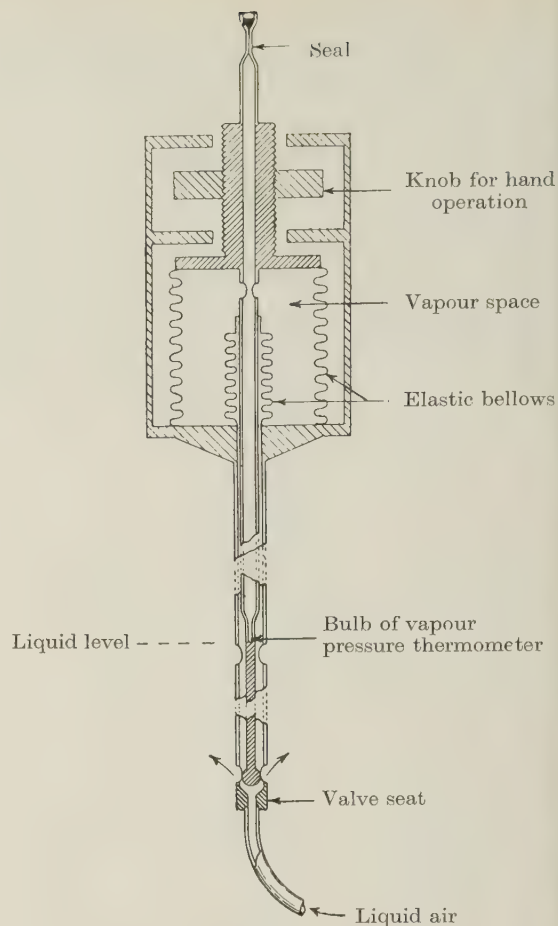
may be used with liquid hydrogen and helium, and have so been used in some laboratories. They are at a serious disadvantage, however, on account of the very low density of these liquids (0.07 g/cc for liquid hydrogen, for instance) and the authors have used another method for liquid hydrogen which is illustrated in Fig. 7. It indicates only when the liquid reaches a predetermined level—a type of indication which is sufficient for many purposes, as when vessels are being filled. The device consists essentially of two open tubes *H*, *J* which sample the fluid continuously at two levels, the lower level being that which is to be indicated and the higher level being one never reached by the liquid. The two samples are drawn through a hydrodynamical “bridge” made up of a differential manometer and four lengths of capillary tubing *A*, *B*, *C*, *D*, two of which are kept at the temperature of the liquid.



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Fig. 7. One-point level indicator

When the liquid reaches the open end of tube *H*, the bridge goes off balance because a greater mass of fluid enters this tube, and the manometer indicates a pressure. It also oscillates rather vigorously on account of the



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Fig. 8. Schematic drawing of an automatic level controller

boiling of the liquid entering the warmer parts of the bridge. In Fig. 7 the samples are shown being drawn through the bridge by means of a vacuum pump. This is not always necessary, however, and in the hydrogen liquefier, where liquid hydrogen boils at about 1.5 atm pressure, this is done simply by connecting the low-pressure side of the bridge to a point at 1 atm pressure in the low-pressure hydrogen return line. A fuller account of this device has been given elsewhere.<sup>(7)</sup>

#### LEVEL CONTROLLERS

It is often necessary to maintain constant the level of low boiling-point liquid in a vessel in spite of a high rate of evaporation, and the automatic level controller shown schematically in Fig. 8 can be used for this purpose. It consists essentially of the bulb of a vapour-pressure thermometer connected through a long tube to the space between two metal bellows. The bulb is located at the level to which the liquid is to be kept, and it carries an extension which operates a valve at a point well below the surface. When the liquid reaches this level the bellows collapse and the valve is closed; if the level drops



precipitately below the bulb, the pressure of vapour rises in account of conduction of heat down the tube, and the valve is again opened and admits more liquid. Fig. 9 shows how such an automatic valve may be fitted in order to keep a constant level in a completely enclosed Dewar vessel in which liquid air is boiling under reduced pressure. Three such controllers are in use on the hydrogen liquefier; provided the vapour-pressure thermometer systems are perfectly gas-tight and no ice is allowed to reach the valves they are perfectly reliable. The accuracy to which the level is controlled depends on conditions within the vessel. Fairly still liquid may be controlled within a few millimetres, and violently

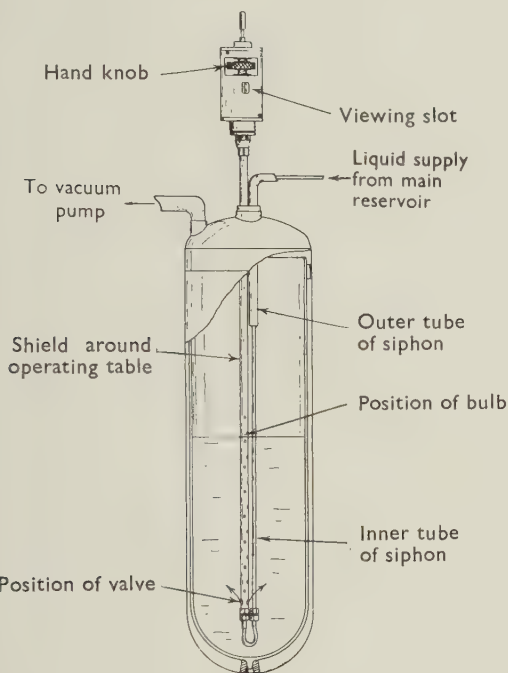
boiling liquid within a few centimetres. The assembly of the double bellows system requires care; a fuller account of the design and method of assembly has been given elsewhere.<sup>(8)</sup> A modified type of controller of simpler design, including only one metal bellows, has also been used. In this case, however, it is not possible to combine the tube leading to the bulb with the spindle operating the valve, as in the device illustrated in Fig. 9.

## ACKNOWLEDGEMENTS

The general scheme for the low temperature installation at the Clarendon Laboratory, Oxford, was due to Prof. F. E. Simon, who has taken a great interest in the work at all stages. A number of acknowledgements have already been made in the text, but we wish to acknowledge particularly the contributions of Dr. N. Kurti, who took part or gave advice in nearly all these developments, of Dr. J. A. W. Huggill, Mr. A. H. Larsen and Dr. F. Mandl, and of the workshop and maintenance staff of the laboratory. Our thanks are due to Mr. W. W. Stonard for Fig. 2.

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Fig. 9. Automatic level controller mounted in an enclosed Dewar vessel

[Paper received 6 February, 1950]

assumptions have been discussed, and the effects of considering conditions which correspond more closely to the experimental arrangement have been examined. Details of the geometry of the focusing system considered here have been given previously.<sup>(1)</sup>

The problem of determining the potential distribution is that of finding the solution of the equation

(a)  $s = 0$  mm; (b)  $s = 1.5$  mm; (c)  $s = 2.8$  mm. Aperture width = 2.54 mm, aperture thickness = 3.18 mm. Anode-cathode distance = 1.3 cm. Filament diameter 0.3 mm



= density per unit volume of space charge in the stem.] As discussed later, the evaluation of the quantity  $4\pi\rho$  presents great difficulty. As a first approximation it has therefore been assumed that the space charge density  $\rho$  is negligible. In this case equation (1) reduces to the simpler form of the Laplace equation

$$\nabla^2 V = 0 \quad (2)$$

of the methods available for the solution of this equation [e.g. relaxation,<sup>(2)</sup> trough,<sup>(3)</sup> or rubber model,<sup>(4)</sup> techniques], that of the electrolytic trough was chosen because it gives sufficiently accurate results rapidly and simply. The experimental details of the equipotential plotting in the present case have already been described.<sup>(1)</sup> The model used represented a section of the system perpendicular to the length of the filament. [A fixed value of the cathode-anode distance,  $d$ , was used, and the subsequent work therefore refers to the system with this value of  $d$ . It is known,<sup>(5)</sup> that the nature of the focal pattern is influenced by the value of  $d$ . These effects, amongst others, will be discussed elsewhere.] Plots were made for various positions of the filament in the cathode. The equipotential maps obtained with the filament at the front, centre, and rear of the cathode are shown in Fig. 1. In the following paragraphs reference is repeatedly made to these three basic filament positions. For the sake of brevity, it is convenient to distinguish them by the distance  $s$  by which the filament is withdrawn behind the plane of the front of the cathode. Thus when the filament is at the front of the cathode (Fig. 1a),  $s = 0$ ; when at the centre (Fig. 1b),  $s = 1.5$  mm; and when at the rear (Fig. 1c),  $s = 2.8$  mm.

At this stage a second assumption was introduced, viz.: that the thermal energies of the electrons leaving the filament were zero. Since the equipotential surfaces close to the filament were circles concentric with the filament, this assumption implied that only electrons leaving the filament radially need be considered. The trajectories of such electrons were obtained by the direct application at each equipotential surface of the relation

$$\sin i = (V_2/V_1)^{1/2} \sin r \quad (3)$$

where  $V_1$ ,  $V_2$  = potentials on either side of the equipotential at which refraction takes place,  $i$  = angle of incidence of electron ray, and  $r$  = angle of refraction of electron ray. Where necessary, additional equipotential surfaces were obtained by interpolation from those plotted experimentally. In all cases plotting was continued until the trajectories were perpendicular to the straight equipotentials in the cathode-anode region; at this stage the paths are parallel to the central axis, and no deviations (due to the equipotentials), can occur. Some typical electron trajectories are shown in Fig. 2.

The electron trajectories themselves merely give the limits of the size of the focal spot. To obtain the distribution of electron density in the focus it is necessary to make some assumption as to the relative numbers of electrons emitted from the filament in different directions.

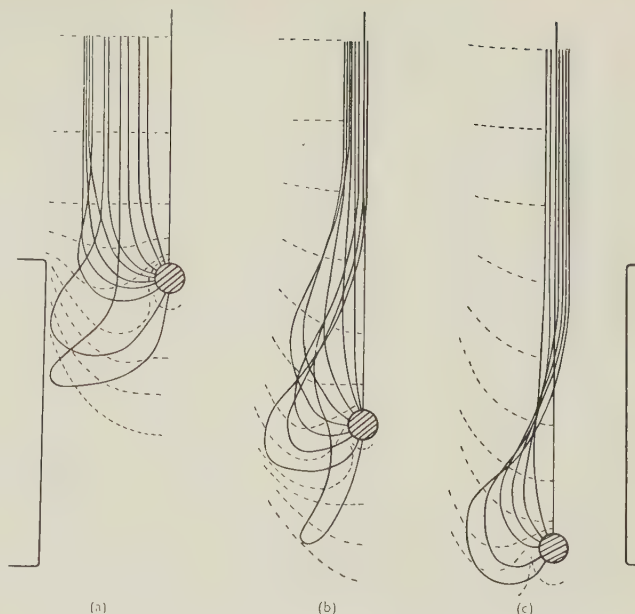


Fig. 2. Electron trajectories in the fields of Fig. 1

(a)  $s = 0$  mm; (b)  $s = 1.5$  mm; (c)  $s = 2.8$  mm

It has been assumed, again as a first approximation, that the same numbers of electrons are emitted in each radial direction. Distribution curves showing the variation of electron density across the focal width were then obtained by measuring the total number of electrons reaching various sections of the focus. These are shown in Fig. 3.

#### DISCUSSION OF RESULTS

*The Distribution Curves.*—A banded focal pattern is obtained at each filament setting. With  $s = 0$  (Fig. 3a), four bands are obtained, the outer pair, at the edges of the focus, being stronger and sharper than the inner pair. With  $s = 1.5$  mm three bands appear (Fig. 3b), one pair at the edges and an equally strong and sharp single band at the centre of the focus. With  $s = 2.8$  mm two bands only arise, as a pair of strong bands at the edges of a focus whose centre is of very weak intensity (Fig. 3c). The overall focal width decreases as  $s$  increases.

The focal patterns obtained for the same filament settings by direct measurement with a pinhole camera are shown in Fig. 4. Comparison of Figs. 3 and 4 shows that the natures of the focal patterns predicted on the simple basis described above correspond closely, qualitatively, to those actually obtained. Consideration of the manner in which the distribution curves are formed, for a wide range of filament settings, shows further that a banded focal pattern, the exact nature of which depends critically on the filament setting, is in general to be expected.

*Origin of Bands.*—The bands in the focal patterns arise as a necessary consequence of the emission of electrons from the filament, in all radial directions, in potential

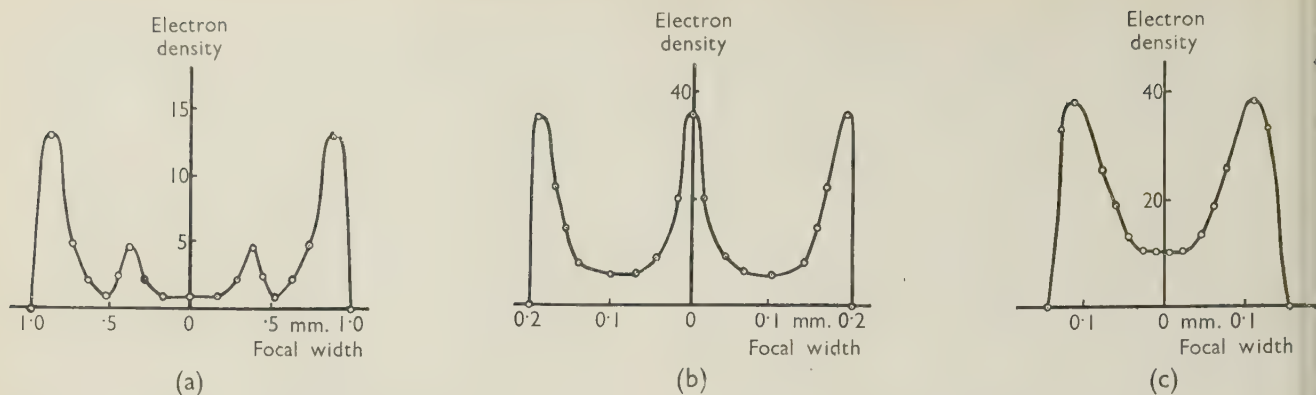


Fig. 3. Distribution curves showing the variation of electron density across the focal width

(a)  $s = 0$  mm; (b)  $s = 1.5$  mm; (c)  $s = 2.8$  mm

fields of the type shown in Fig. 1. At any given filament setting contributions to the electron density in each band are made by electrons emerging from the filament at different angles to the central axis. Such electrons, of course, originate at different parts of the filament surface. In any particular focal pattern each band is composed of electrons emitted from the filament in several different angular ranges. The occurrence of the bands is not directly attributable to a simple mechanism, such as that, suggested by Beese,<sup>(6)</sup> of emission from particular areas of the filament.

A variation in the filament setting causes a change in the position and extent of the region within the focal pattern to which electrons emitted in a given angular range are focused. That is, a change in  $s$  modifies the potential field and alters the trajectories of electrons emitted at any given angle to the central axis. The consequent re-grouping of electrons emitted in different angular ranges results in a change in the distribution curve. On this basis the observed variations of the focal patterns with the filament setting (see reference<sup>(1)</sup>), can be qualitatively explained.

**Rounded Appearance of the Bands.**—A second characteristic of the foci obtained in the present system is also illustrated in Fig. 4. This is the rounded appearance of the bands in the focal patterns.

When  $s$  is small the focus is four banded with the outer pair of bands slightly convex and the inner pair concave. This is an effect occurring in a direction parallel to the length of the filament. However, a simple additional assumption enables the foregoing results (derived for a section perpendicular to the length of the filament), to give an explanation of this phenomenon. This assumption is that the filament bows slightly, bringing the centre closer to the front of the cathode than are the ends. The nature of the line filament justifies this assumption. There may be (a) an initial bending resulting from the shaping of the filament from a straight wire into a flat "U," (b) bending due to the way in which the filament is held in its supports, and (c) addi-

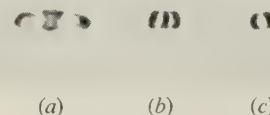


Fig. 4. Pinhole photographs of the focus.

(a)  $s = 0$  mm; (b)  $s = 1.5$  mm; (c)  $s = 2.8$  mm. Focal width along the horizontal, foreshortened focal length along the vertical.

tional bending caused by subsequent thermal expansion of the filament. The displacement of the centre of the filament relative to the ends need only be approximately 0.2 mm, since in this region the curve of focal width against filament setting has a very steep slope. For further details see<sup>(1)</sup>. If the length of the filament is then considered as composed of many short sections, the electron trajectories and distribution curve for each section can be obtained as outlined above. For the central section these are as shown in Figs. 2a and 3a. Here all electrons from the range  $\pm 65^\circ$  approx. are included in the inner bands and the central region of the focus, while the majority of the remainder form the outer bands. For sections nearer the ends of the filament re-grouping occurs. Electrons from the same range,  $\pm 65^\circ$ , are now spread over a wider region, with a consequent increase of the inner band separation; the overall focal width is simultaneously reduced. Superposition of the patterns for the various sections shows that the outer bands should be slightly convex and the inner concave.

As  $s$  is increased the bands should first tend to straighten, since the re-grouping produced by the small change in setting between the various sections is less pronounced, cf. Fig. 4b. Subsequently the bands should re-assume a convex shape because of a superposition effect similar to that discussed above, cf. Fig. 4c. (The changes from the four- to two-banded focal patterns occur simultaneously and independently.) Referring to the electron trajectory maps of Fig. 2, it may be noted



at the three cases correspond approximately to regions of filament setting for which (a) no electrons cross the central axis, (b) some electrons forming the outer bands cross the axis, and (c) almost all electrons cross the axis. The appearance of the bands can also be correlated with the regions of the curve of focal width against filament setting (see reference<sup>(1)</sup>, Fig. 4). At small values of  $s$  the steep negative slope of the upper curve corresponds to convex outer bands and the positive slope of the lower curve to concave inner bands. For intermediate values of  $s$  extending over the broad maximum and minimum respectively on the separate curves the small slopes correspond to small band separations. At larger values of  $s$  the larger negative slope again corresponds to convex bands.

**Quantitative Estimates of Focal Dimensions.**—The band widths and spacings obtained in practice are always found to be greater than those predicted from the distribution curves. A comparison of the observed and predicted focal dimensions is made in Table 1. Here  $d_1$  and  $D_1$  denote the observed and predicted outer band widths and spacings, and  $d_2$  and  $D_2$  the corresponding inner band widths and spacings [when  $s = 1.5$  mm  $d_2$  and  $D_2$  are taken as the central band width]. Although great accuracy in plotting the electron trajectories and in deriving the distribution curves is not claimed, the order of magnitude and general trend of these figures are considered significant. Both the ratios  $d_1/D_1$  and  $d_2/D_2$  increase as  $s$  increases. The large value of  $d_2/D_2$  obtained when  $s = 1.5$  mm is particularly to be noted.

Some limitations of the simplifying assumptions made in deriving the distribution curves are now considered, and the modifications introduced by making allowance for the thermal energies of emission of electrons from the filament and for the effects of space charge are discussed.

#### THERMAL ENERGIES OF EMISSION

The Fermi-Dirac distribution of energy for electrons emerging from a hot filament approximates very closely to a Maxwellian law. Electrons leaving the filament have velocity components both normal to the emitting surface and in a plane perpendicular to this direction. Corrections to the previous derivation of the electron density distributions are therefore necessary both in respect of the velocity of emission and because the emission can no longer be considered as limited to purely radial directions. The velocity distribution of electrons emerging normal to the emitting surface can be expressed by a relation of the form

$$dN_z = (Nm\dot{z}/KT)[\exp(-m\dot{z}^2/KT)]d\dot{z} \quad (4)$$

where  $dN_z$  = number of electrons emitted per unit area per unit time with velocity components along the  $z$  axis (normal to the surface) between  $\dot{z}$  and  $\dot{z} + d\dot{z}$ ,  $m$  = electronic mass,  $K$  = Boltzmann's constant,  $T$  = absolute temperature of emitting surface.<sup>(7)</sup> The evaluation of this relation shows that, in the present system, some 80% of the electrons emitted normal to the surface have velocities equivalent to less than 0.5 volt. A similar expression, yielding the same numerical values, holds for the velocity distribution in the plane perpendicular to  $z$ . This thermal energy only becomes comparable with the potential due to the applied h.t. voltage in the immediate vicinity of the filament, and, in some cases, for an additional limited region in the cathode. Between the cathode and anode the thermal energy is negligible compared with the potentials occurring along the electron path. Fig. 1 shows that the axial potential in the plane of the cathode surface is still approximately 1%, i.e. 500 volts for an applied voltage of 50 kV. The effects of these thermal energies have been estimated by the following method, which obviates the necessity for an exact calculation of the ranges of velocity and direction of emission within which any known percentage of the emitted electrons are included. At each point on the filament surface, at which previously a single radial ray of zero energy electrons was supposed to have emerged, electrons leaving at angles up to  $90^\circ$  to the normal and with energies of 0.5 volt have been considered. It is found that large changes are caused only in the initial parts of the trajectories. The total spread about the position of the normally emitted ray of zero energy is of the order of a band width (i.e. approximately 0.07 mm), corresponding to an increase in overall focal width of about 20%. In general, thermal effects tend to broaden the bands in the focus and so increase the predicted focal dimensions, but the scale of the effect is far smaller than necessary to explain the discrepancy between the observed and predicted values of the focal widths and band spacings.

#### SPACE CHARGE EFFECTS

The second assumption made in deriving the electron trajectories was that no space charge was present. As stated earlier, this assumption was made rather for the purposes of simplification than as representing the true state of affairs. Space charge would affect the results given above in two ways, one due to the mutual repulsion of the electrons in the beam, the other due to the changes set up in the potential distribution. These effects are now considered separately.

Table 1. Comparison of predicted and observed focal dimensions

Filament setting $s$ (mm)	Observed spacings (mm)		Predicted spacings (mm)		$\frac{d_1}{D_1}$	$\frac{d_2}{D_2}$
	Outer $d_1$	Inner $d_2$	Outer $D_1$	Inner $D_2$		
0.0	7.3	1.2	1.7	0.7	4	2
1.5	3.5	0.4	0.4	0.02	9	20
2.8	2.8	—	0.2	—	14	—

**Mutual Repulsion.**—In the space charge theory applied by Klemperer<sup>(8)</sup> to strip emission systems the beam spread caused by mutual repulsion in a path of length  $z$  is given by a term of the form  $(1/4k) (I/V^{3/2}) z^2$ . Here the forward direction of motion of the electrons is along the  $z$  axis, and the spread occurs in the  $y$  direction.  $I$  is the line current per unit length in the  $x$  direction (perpendicular to the  $yz$  plane),  $V$  is the potential to which the electrons are raised, and  $K$  is a constant.  $K = 10.4$  if  $I$  is measured in microamps/cm length. This relation holds for constant velocity electrons, provided that the problem can be treated as two-dimensional. In the present case difficulties arise both because the electron path lies wholly in an accelerating field and because the current distribution across the beam is non-uniform. These have been met by utilizing a method of numerical integration, applicable to each band separately. An expression giving the order of magnitude of the band spread,  $\Delta y$ , is

$$\Delta y = \sum_{V_1} \frac{1}{4k} \left[ \frac{f(I_1)}{V_1^{3/2}} \right] \delta z_1^2 \quad (5)$$

Here  $\delta z_1$  = length of path over which the potential (assumed constant), is  $V_1$ ,  $f(I_1)$  = current per unit length in potential region  $V_1$ , and the summation extends over the whole electron path. The current function  $f(I_1)$  was introduced because the band width is not constant at all points along the electron path. The relation used to give approximate values of  $f(I_1)$  was

$$f(I_1) = I \frac{w_0}{w_1} \quad (6)$$

where  $I$  = total current per cm length in the band,  $w_0$  = width of band between 10% equipotential and anode,  $w_1$  = width of band in potential region  $V_1$ . (This method of referring  $I$  to a beam of width  $w_0$  instead of to a beam converging to a focus of zero width tends to overestimate the spread due to mutual repulsion.) In this relation the values of  $I$ ,  $w_0$ , and  $w_1$  were obtained from the appropriate set of electron trajectories. The steps of voltage ( $V_1$ ) used were the same as those employed in deriving the electron trajectories, and the values of the potential were taken from the trough measurements. Results have been calculated for a filament setting of  $s = 1.5$  mm. Values of anode voltage and total beam current of 50 kV and 20 mA respectively were assumed.

The space charge factors  $\left[ \frac{f(I_1)}{V_1^{3/2}} \mu\text{A/cm/volts}^{3/2} \right]$

vary widely along each mean trajectory.<sup>(8)</sup> Some typical orders of magnitude are given in Table 2. Near the filament the space charge factors may be very large; their effect is, however, offset by the short lengths of the mean electron paths in this region. Between the 10% equipotential (where the equipotentials become straight lines and the trajectories are all parallel to the central axis) and the anode the calculated spread for each band is approximately  $2 \times 10^{-3}$  mm. This is negligible compared with the predicted band width of  $7 \times 10^{-2}$  mm. Thus, so far as mutual repulsion is concerned, the focal pattern derived at the 10% equipotential fairly represents that obtained on the anode. Between the filament and the 10% equipotential the spread for each outer band is approximately 0.06 mm. For the inner band, however, the corresponding spread is approximately 0.2 mm. As noted previously, this inner band is the one for which the large differences between the observed band width,  $d_2$ , and the band width predicted on the simple assumptions,  $D_2$ , give rise to the significantly large value of  $d_2/D_2$ . This large increase in the spread arises because electrons here not only pass through much weaker fields than in the former case, but also have longer paths in these weak fields. The overall focal width is thus only slightly increased, though the central band may be considerably widened. On a similar basis it is found that mutual repulsion effects will be negligible when  $s = 0$  mm, but that when  $s = 2.8$  mm a spread of each band of approximately 0.2 mm is probable. Mutual repulsion effects, like those of thermal energies, tend to produce a broadening of the bands and an increase in overall focal width, but the effect is again on too small a scale to account for the difference between the observed and predicted results.

**Potentials Induced by Space Charge.**—When allowance is made for the presence of space charge the potential at any point in the inter-electrode region is not that given by the trough measurements, but is modified both by the potential at that point due to the beam itself, and by the potential due to the charges induced by the beam on the surrounding electrodes. The electron trajectories, and the distribution curves, are consequently changed. A thorough quantitative examination of these effects would be complex and lengthy, and has not yet been attempted. Some general ideas can, however, be fairly readily obtained, though they are only to be regarded as tentative. (It should be noted that, since quite good qualitative agreement is obtained on the simplest basis, the mechanism postulated to account

Table 2. Typical space charge factors along electron path when  $s = 1.5$  mm

Equipotential (%)	100	10	1	0.1	0.01
Space charge factor					
$\left( \frac{f(I_1)}{V_1^{3/2}} \right) \mu\text{A/cm/volts}^{3/2}$	$8 \times 10^{-3}$	$2 \times 10^{-1}$	9	$3 \times 10^2$	$8 \times 10^3$



the quantitative difference should magnify the focus without radically altering the current distribution within

As a result of space charge the curvature of the equipotentials within the cathode will tend to be increased, and that of those between the cathode and anode reduced.<sup>(9)</sup> Reference to Figs. 1 and 2 shows that both factors would tend to produce changes in the electron trajectories which would increase the focal dimensions. Very approximate calculations show that this increase may be on a scale comparable with the observed discrepancy. Since the change in potential distribution will affect all electrons and not only those emerging from the filament in individual angular ranges, only a slight regrouping effect would be expected. A greater increase in focal dimensions would be obtained with larger values of  $s$  for two reasons. First, the magnitudes of the potentials near the filament (due to the applied h.t. voltage), are decreased, making the effect of the potentials arising from the space charge relatively more important. Second, the electron paths in these disrupted fields are longer. This agrees with the result that the ratios  $d_1/D_1$  and  $d_2/D_2$  increase as  $s$  increases (Table 1).

#### DESIGN OF FOCUSING SYSTEMS

Both the pinhole camera and electrolytic trough results show that with a focusing system of the type described above a banded focal pattern is always obtained, except in the extreme case of  $s$  so great that negligible current is obtained. This non-uniformity of the X-ray source may lead to unnecessary complications in many X-ray techniques, and usually severely restricts the possible loading on the target. For the maximum permissible target loading on the area of a band is often exceeded long before the total loading over the whole focal area reaches the limiting value. This causes a reduction in the possible X-ray output and an increase in exposure times. To achieve uniformity of electron density over the focus it would seem necessary that the filament and cathode were designed as a unit to meet the electron-optical requirements. In this connexion the possibility seems to be the use of some form of strip filament. Since almost the whole of the focusing action occurs within or very near to the cathode, the exact

form of the potential field between the cathode and anode is of secondary importance.

The common method of describing focal patterns in terms of a "central focus with a penumbra" is most misleading. The "penumbra" is an integral part of the focus and cannot be considered as separate. Changes in the focal pattern caused by geometrical alterations or the use of bias,<sup>(5, 10)</sup> then act on the focus as a whole. The resulting focal pattern may, or may not, be equivalent to the removal of the "penumbra," but the mechanism of the change is not that of simple subtraction of a part of the original focal pattern.

#### ACKNOWLEDGEMENTS

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# Electrical Conductivity of Oxide Cathode Coatings

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Conductivity measurements on activated coatings give results similar to those of Vink and Loosjes. Their theory implies barium concentration of at least  $10^{19}$  atoms/cm<sup>3</sup>. Hall effect measurements indicate  $10^{14}$  electrons/cm<sup>3</sup> at 1 100° K, and free path  $2 \times 10^{-5}$  cm. This result may not in fact be inconsistent with the Vink-Loosjes theory. P-type conductivity is not encountered in well activated coatings, and is a feature developing while drawing emission under non-activating conditions. It is due to oxidizable impurities in the coatings. Rapid poisoning or activating effects at low temperatures emphasize the importance of phenomena at surfaces, and it is suggested that these are associated with electron or hole movement rather than ion movement.

The experiments<sup>(1)</sup> on the conductivity  $\sigma$  and the Hall effect  $R$  in oxide coatings sprayed on a magnesia base have been continued. The base was provided with contacts formed by firing on, in hydrogen, suitably shaped patches of powdered molybdenum, followed by powdered nickel in which leads were embedded. The magnesia was in the form of a rectangular plate, with two end and two side contacts. The coating was sprayed on one side of the plate to a thickness of 0.1 mm, covering the contact patches. The other side of the plate was in contact with an alumina-coated heater spiral. Preliminary experiments established the behaviour of the uncoated magnesia. A tantalum anode facing the coating enabled emission to be studied.  $R$  and  $\sigma$  have again been calculated by taking the observed thickness of the oxide layer, not corrected for density, which lay between 1.0 and 1.4.

## THE REPRODUCIBLE STATE

With any type of coating after outgassing at 1 300° K, a state I is at once established (Fig. 1) where  $\sigma$  is near  $10^{-3} \Omega^{-1} \text{cm}^{-1}$  at 1 100° K, and where the slope of  $\log \sigma$  against  $1/T$  is about 1.1 eV. There is surprisingly little variation in these figures from one sample to another, and moreover it is this state to which the coating reverts after either poisoning in an oxidizing gas or activating in a reducing gas. After poisoning, recovery to state I occurs on running in vacuum at 1 100–1 300° K, while after activating, there is a slow return to state I, on running, which above 1 050° K can be due to barium evaporation, or at lower temperatures, to poisoning. The conductivity in state I is wholly N-type, at least down to 650° K; taking the Hall effect formula for a homogeneous semi-conductor, we find that the density of free electrons  $n$  at 1 100° K is near  $5 \times 10^{12}/\text{cm}^3$ , the free path is  $3 \times 10^{-5}$  cm, and the slope of  $\log n$  against  $1/T$  is 1.2–1.3 eV. This is the same as the slope of  $\log I$  against  $1/T$ , where  $I$  is the emission, and is slightly higher than the slope of  $\log \sigma$ . When poisoning occurs,  $n$  and  $I$  fall by precisely corresponding ratios, and the fall in  $\sigma$  is similar. The slopes are all steeper, and for example if  $n$  and  $I$  fall by a factor of 100 at 1 000° K the slope of each increases to about 2 eV.

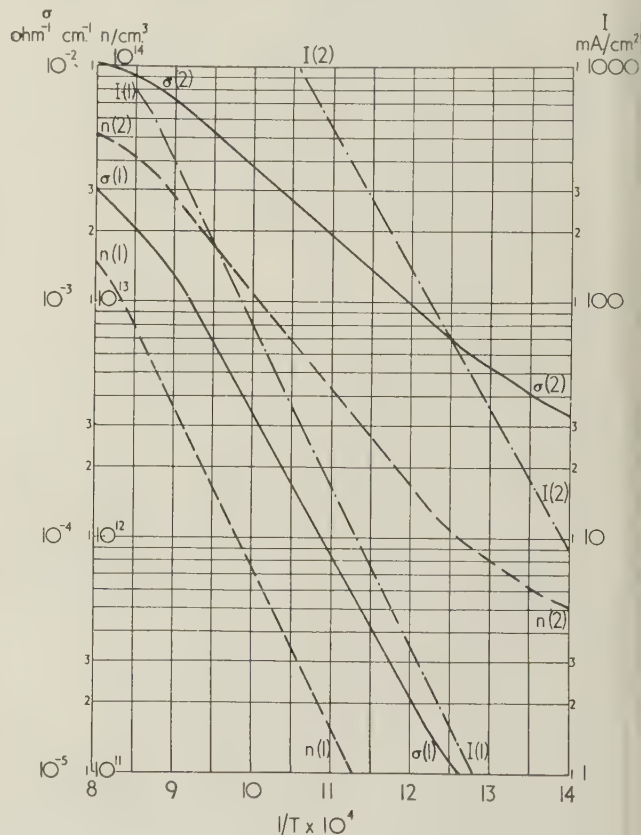


Fig. 1. Relation between conductivity  $\sigma$ , emission  $I$  and  $1/T$ .

This state I has been encountered by many previous observers,<sup>(2, 3, 4)</sup> but the points to be emphasized are: (a) its ready reproducibility; (b) that while it is by no means an activated state, neither is it a "poisoned" one; it is in some sense a typical condition of an oxide coating, and as such it is unlikely to be dependent on details of concentration of barium atoms or other centres either in crystals or on surfaces; and (c) the relation between  $I$  and  $\sigma$  is different in states which are poisoned relative to state I, compared with states activated with respect to state I. It is only in poisoned states that we find that in any state, the slopes of  $\log I$  and  $\log \sigma$  are similar over a range of temperature. On activation, though  $I$  at 1 000° K may increase by a factor of 100 or more



mpared with state I, the work function, or the slope of  $I$  against  $1/T$ , decreases only by 0.1 or 0.2 eV. On the other hand, the slope of  $\log \sigma$  near 1000° K only decrease from 1.1 eV to 0.5 eV; in addition, at lower temperatures we observe a still lower slope, about 0.2 eV, and it is clear, therefore, that the simple model of a homogeneous semi-conductor is not applicable. According to this model, in Fig. 2, where  $\epsilon$  gives the slope of  $\log n$ , the work function is given by  $\chi + \epsilon$ ; this could apply in our poisoned states, and has been observed to apply over a range of activation by Eisenstein,<sup>(5)</sup> by Hannay, McNair, and White,<sup>(4)</sup> and by Nishibori and Kawamura.<sup>(6)</sup> It cannot apply, however, in the really well-activated states; this point is a very important one, and is discussed below.

#### LOSS OF ACTIVATION

It should be noted that in order to obtain good activation of coatings on magnesia, we have had to work with reducing atmospheres. Most of this work has involved treatment in ethane with the cathode at 800–1100° K. This has given values of  $\sigma$  near  $10^{-2} \Omega^{-1} \text{cm}^{-1}$  at 100° K—state II. In this state the slope of  $\log \sigma$  is 0.5–0.7 eV at 1000° K, and 0.2 eV below 800° K,  $n$  at 100° K is about  $10^{14}/\text{cm}^3$ , and the conductivity is again wholly N-type at least down to 650° K. After ethane activation, running above 1050° K causes a drop in  $\sigma$ , and  $n$ , until finally state I is restored. Since Hannay, McNair and White always heated above 1300° K after ethane treatment, they were able to observe only a state similar to our state I. This high temperature loss of activation is due to disappearance of active centres, perhaps by evaporation, and causes a temperature hysteresis, together with considerable uncertainty as to the true slope of  $\log \sigma$  or  $\log n$  above 1100° K. In addition to these effects, operation below 800° K causes a slow return to state I; a drop of this kind can occur at room temperature, which indicates that the activation cannot have been due to formation of carbon, but rather to barium, which is then destroyed on exposure to the pump vacuum at low temperature; the vacuum was about  $8 \times 10^{-7}$  mm of mercury. During attempts to activate by drawing emission, such high values of  $I$ ,  $n$  or  $\sigma$  have not been reached as we obtained after ethane activation, but the effect on slopes has been of the same type, and the loss of barium above 1050° K and the poisoning below 800° K have occurred in the same way.

#### P-TYPE BEHAVIOUR

If a coating is sealed off from the pump in state I, and activated partially by drawing emission, a state intermediate between I and II is achieved. During operation at 1000–1100° K, taking emission, a new feature is observed, in all samples, in that while  $\sigma$  falls, the low temperature conductivity becomes P-type. A P–N transition is detected first near 800° K, and moves

to higher temperatures as operation proceeds. It finally stabilizes after a few tens of hours, at a temperature which for Ba/SrO is usually between 1000 and 1050° K, but is normally at lower temperatures for Ba/Sr/CaO, and higher for BaO. Finally  $\sigma$  has lower values than in state I, and  $I$  is considerably lower than after the activation. If, at any time during this operation, an activating treatment is given, the transition is moved to lower temperatures, and  $I$ ,  $n$  and  $\sigma$  are raised, but during non-activating runs, all three fall again. This is the behaviour previously described,<sup>(1)</sup> and the behaviour at the transition indicates two types of conductivity in parallel, one P and one N. The following further facts have been established since the earlier communication:<sup>(1)</sup> (a) the transition does not appear during operation on the pump; (b) it is not present in a really well activated coating, e.g. in state II; (c) if, after it has appeared and stabilized, in a sealed off tube as described, the tube is opened to the air and re-evacuated, the transition has gone, and the conductivity is wholly N-type again; (d) it does not reappear either on the pump or after sealing off a second time, and, moreover,  $I$ ,  $n$  and  $\sigma$  are better maintained than in the first run. It is clear that the substance causing the hole conductivity is foreign to the Ba/SrO system; in particular it is not due to barium deficiency in the crystals, which was a remote possibility.<sup>(1)</sup> Since it is so readily oxidizable, it may well be due to alkali metal; for example  $\text{K}^+$  ions replacing  $\text{Sr}^{++}$  would give hole conductivity. Evidently the formation of this substance comes to an end point in the first run, and it is not reformed following oxidation. It is natural to speculate whether this substance is initially an alkali metal chloride, causing the well-known liberation of chlorine. The important point is, however, that this effect is definitely foreign to the Ba/SrO system, and is not present in well-activated coatings.

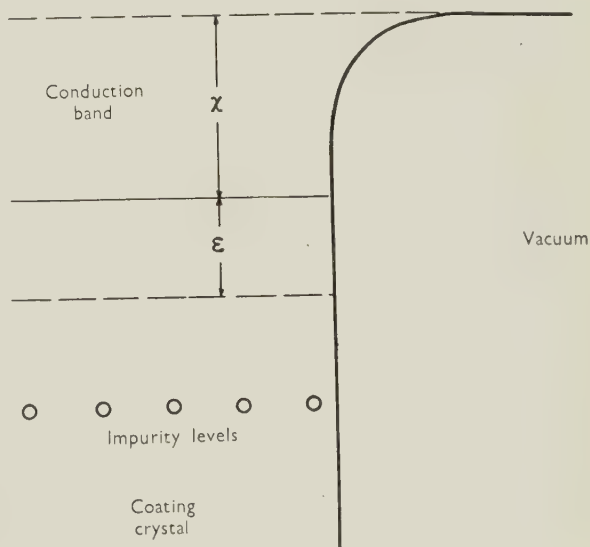


Fig. 2. Energy levels

## THE VINK-LOOSJES MODEL

We have shown above that the model for a homogeneous semi-conductor does not apply to well-activated coatings, where  $\epsilon$  continues to decrease with little change in work function. There are no doubt several possible explanations of these facts, but two in particular claim attention. It might be that thermionic emission arises by thermal ionization of adsorbed barium, as in de Boer's model, so that work function is not related to the values of  $\chi$  or  $\epsilon$  for the crystals, when such barium is present. There is associated with this the possibility that in poorly activated states, no surface barium is present, and the semi-conductor model does apply to the "clean" crystals, whereas when surface barium is present in well-activated states, this model no longer applies. This idea has been suggested previously by the writer,<sup>(2)</sup> where it was thought that d.c. operation might remove surface barium and give a "clean" surface. The other theory is that of Vink and Loosjes,<sup>(8,9)</sup> which states that conductivity is due to two components, one true crystal conductivity through contacts, and the other a parallel conductivity through the space charge in the pores in the coating. It will be realized at once that this model is in qualitative agreement with our observations, since it would require conductivity in poisoned states to be determined by emission between crystals; here we found the same slopes for  $\log n$  and  $\log I$ ; whereas in activated states the emission and the conductivity of the crystals both increase, and the two effects in parallel can give rise to the observed curves for  $\log \sigma$ . It should be noted that Vink and Loosjes' results also showed that the slope of  $\log \sigma$  continued to decrease with little change in work function, as in our observations.

The important feature of the Vink-Loosjes model is that the slope of  $\log \sigma$  for the crystals is the low-temperature slope, and that this slope is the value we should take for  $\epsilon$ . Both this value of  $\epsilon$  and the change in this value during activation are smaller than corresponding values for the slope at 1000° K, previously assumed to correspond with  $\epsilon$ . Incidentally these lower values of  $\epsilon$  are much more probable than values near 1 eV, since the ionization potential of barium is not very high, and the dielectric constant is at least 14 in static measurements and 4 at infra-red frequencies. Taking these low temperature values for  $\epsilon$ , it is now possible for the semi-conductor model to apply again. This can be seen from Table 1 in the Vink-Loosjes paper,<sup>(8)</sup> where if we subtract values of  $\epsilon$  from the work function, we get a fairly constant quantity, which from Fig. 2 we identify with  $\chi$ . Thus with this model there is no need to postulate surface barium, but it is supposed that without adsorbed barium,  $\chi$  lies between 0.7 and 1.0 eV, and  $\epsilon$  between 0.5 and 0.1 eV according to the degree of activation. The values of  $\sigma$  and  $I$  would then both depend on  $n_0^{\frac{1}{2}}$ , where  $n_0$  is the density of excess barium, present either interstitially or as equivalent oxygen ion vacancies.

Another important feature of this low value of  $\epsilon$  is

that although the slope is so low,  $\sigma$  is still rising with temperatures up to 800° K or more. With  $\epsilon$  0.2 eV or less, complete ionization of the impurity centres would be expected, and with it a maximum in  $\sigma$ ,<sup>(10,11)</sup> at temperatures below 800° K, unless  $n_0$  is greater than  $10^{19}/\text{cm}^3$ . Values at least of this order are therefore required to fit this model. Some earlier measurements on free barium were of this order, while although the recent measurements of Jenkins and Newton<sup>(12)</sup> in their laboratory give lower figures, the present work with continuously pumped systems indicates that their cathodes may not have been fully activated; they had similar vacuum systems, and after activation the cathodes were exposed to this "vacuum" at room temperature. Emissions ten times greater than their emission are common, which might correspond with  $n_0$  a hundred times greater, so that this work is not necessarily inconsistent with the Vink-Loosjes model.

A further point is that with  $n_0$  as high as this, with  $\epsilon \sim 0.2$ ,  $n$  would be expected to be of the order of  $10^{17}$ – $10^{18}$ . This contrasts with our Hall effect measurements giving  $10^{14}$  at 1100° K in our state II. It is realized that under the conditions of the Vink-Loosjes model, a new analysis is necessary to determine the true significance of a Hall effect measurement. It is unlikely, however, that any large error is involved in estimating an effective value of  $n$ ; the question is the significance of this effective value. It should be noted that the mean space charge density in the pores is given by Vink and Loosjes as  $(2\pi m/kT)^{\frac{1}{2}}I$ , which with  $I = 1 \text{ A/cm}^2$ , is of the order  $6 \times 10^{12}$  electrons/cm<sup>3</sup>. If it is this density which is the effective one, rather than that in the crystals, our results may again not be inconsistent with this model. This point will require further consideration.

As regards emission, the semi-conductor formula is:<sup>(13)</sup>

$$I = DA_0 T^2 n_0^{\frac{1}{2}} \frac{h^{3/2}}{(2\pi m k T)^{\frac{3}{2}}} \exp(-\phi/kT)$$

With  $n_0 = 10^{19}$ , as the Vink-Loosjes theory seems to require, and with  $D = 1$ ,  $A_0 = 120$ ,  $\phi = 1.1 \text{ eV}$ , the emission  $I$  is 300 A/cm<sup>2</sup> at 1000° K, which is much higher than observed. There are many indications, however, that the true work function near 1000° K is about 1.5 eV, not 1.1 eV. With  $\phi = 1.5 \text{ eV}$ ,  $I$  is about 10 A/cm<sup>2</sup>, which agrees with observation for the best states of activation.

The alternative formula for fully ionized donating centres is:<sup>(14)</sup>

$$I = Dn_0 \epsilon \sqrt{kT/2\pi m} \exp(-\phi/kT)$$

which would give with  $D = 1$ ,  $\phi = 1.1$ ,  $n_0 = 10^{19}$ , an emission  $I$  of 30 A/cm<sup>2</sup>. This would also be greatly reduced if  $\phi$  is 1.5 eV. The observed values of emission cannot therefore be used as an argument for or against Vink-Loosjes theory.

## PROCESSES AT SURFACES

Thus the results first described above are in accordance



th the Vink-Loosjes model at least qualitatively, and the results described later are not necessarily inconsistent with it. Mention must be made of some other experimental results obtained, which stress the importance of surface effects, and which have previously been considered to support the first model referred to above. Thus poisoning due to oxygen occurs "instantaneously" on introducing oxygen not only at 1 100° K and 800° K, but also at 675° K. It seems unlikely for diffusion in thin crystals to occur so rapidly at so low a temperature, and we consider this a surface effect. Another point follows from a series of experiments on special diodes with tungsten anodes, which were outgassed at 2 600–300° K, the cathodes being moveable, away from the anodes, during their outgassing. Here good emission is obtained at once after sealing off without activation, and, moreover, the d.c. emission was exactly equal to the pulsed emission at all temperatures where both could be measured. With a hot cathode and cool anode, deposits form on the anode, lowering the d.c. emission, but not the pulsed. On cleaning the anode at 2 600° K, the d.c. emission is restored. Thus it is conclusive that the two types of emission are essentially the same, and the d.c. emission is normally lower than the pulsed due to poisoning effects. These occur within  $10^{-3}$  sec.<sup>(13)</sup> It is thus probable that surface effects are probably predominant. Since these poisoning processes affect  $\sigma$  as well as  $I$ , there is perhaps some further support for the Vink-Loosjes model, since rapid effects on the emission component  $\sigma$  are more probable than rapid effects on the true crystal conductivity.

On the other hand, it is believed there is another possible interpretation of surface effects. It is suggested that following carbonate decomposition, lattice defects are frozen in on cooling, according to the model of Iijboer,<sup>(15)</sup> and that there are many defects without associated electrons. The change of slope near 1 200° K is not considered to be associated with this, as Eisenstein suggested,<sup>(5)</sup> but nevertheless it is believed that this does occur. Then the effect of contact with oxidizing or reducing agents is not necessarily to cause true chemical changes with associated movements of ions, but much more simply to subtract or add electrons, via the surface of crystals, to the centres which were frozen in. In this way the rapid poisoning or activation at low temperatures which have been observed might be explained. This would, moreover, predict that in a single crystal, with fewer frozen in defects, activation might be impossible, even in methane, as observed by Sproull.<sup>(16)</sup> It would also predict that the ultimate state of activation should depend on the maximum temperature of processing, for which there is a certain amount of experimental support. It might even be an important factor in determining the advantages of mixed oxides over the single oxide BaO, while yet another point is that assuming the surface process is one of adsorption, the surface/volume ratio would be one of great importance, indicating the need for small coating particles.

## CONCLUSION

Our conductivity measurements are readily explained in terms of the Vink-Loosjes model, but not by a homogeneous semi-conductor model. Results on barium content of coatings, on emission, and the Hall effect measurements are not directly in support of the Vink-Loosjes model, though there is possibly no real inconsistency. The part which may be played by surface adsorbed barium remains obscure; this is not a necessary feature of the Vink-Loosjes theory, which allows a semi-conductor model to fit the observations. Surface barium may, however, be a direct source of emission, explaining the increase in emission with small decrease in work function, and the difference between pulsed and d.c. performance.

It occurs to the writer that a layer of surface barium could be the source of the low temperature conductivity, which varies only slightly with temperature, as observed in highly activated states in the present experiments and in those of Loosjes and Vink. This conductivity would be in parallel with that through the coating, which varies more steeply, and which overwhelms the surface conductivity above 800° K. Alternatively, the effect of surface barium, or of other adsorbed layers, may be to modify the electron concentration in the conduction band because frozen-in defects are present in the crystals. It is hoped to carry out further experiments to decide between these various possibilities.

## ACKNOWLEDGEMENTS

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# A Method of Locking Oscillators in Integral and Non-Integral Frequency Ratios

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The output voltages of two audio oscillators required to operate in an integral or non-integral frequency ratio are applied to a non-linear mixer. The appropriate zero beat is used to provide a suitable correction voltage to a frequency determining circuit forming part of one of the oscillators. A practical arrangement is described by which an  $R$ - $C$  oscillator may be locked with a 1 000 c/s maintained fork in quite high orders of frequency ratio (e.g. 17 : 7). Further development of the principle is discussed.

Since Appleton<sup>(1)</sup> in 1922 first gave the matter careful theoretical study, the problem of controlling or "locking" one oscillator with another has received extensive treatment in the literature. Van der Pol and Van der Mark<sup>(2)</sup> discovered that a relaxation oscillator of natural frequency close to  $f/n$ , where  $n$  is an integer, could be made to operate at precisely that frequency by injecting a voltage of frequency  $f$ . A generator of standard audio frequencies based on this principle and utilizing multi-vibrators was described by Essen<sup>(3)</sup> in 1936.

The presence of a non-linear conductance or process in the oscillator circuit is an essential feature of the locking phenomena considered in these and a large number of other papers. It is not, therefore, surprising that the inherently non-linear relaxation oscillator has generally proven most suitable for frequency division. The special "quasi-stable" circuits described by Sterky,<sup>(4)</sup> Miller<sup>(5)</sup> and Fortescue<sup>(6)</sup> are, however, exceptions. The usefulness of the multivibrator has been further extended by Davis<sup>(7)</sup> to obtain non-integral rational ratios.

Recently an urgent need arose for a sine-wave oscillator having a frequency stability of the order of a few parts in  $10^6$  and capable of operating at a discrete set of frequencies in the 1–3 kc/s range with a spacing not greater than about 3% (continuous tuning was not necessary). Since the only suitable standard readily available was a 1 000 c/s maintained fork, a method of locking in non-integral ratios was demanded. The following device, developed independently by the author, appears to be similar in principle to a method of synchronization described by Tucker.<sup>(8)</sup>

Most systems of synchronization have in common the injection of the control tone into the oscillator circuit, but Tucker showed that comparison of the control tone and that of the free-running oscillator might be carried out separately and a suitable correction applied to the oscillator frequency. This principle need not, however, be confined to direct synchronization. Indeed it appears to possess outstanding advantages when it is desired to control a sine-wave oscillator at a multiple or sub-multiple of the controlling frequency or in a non-integral rational ratio.

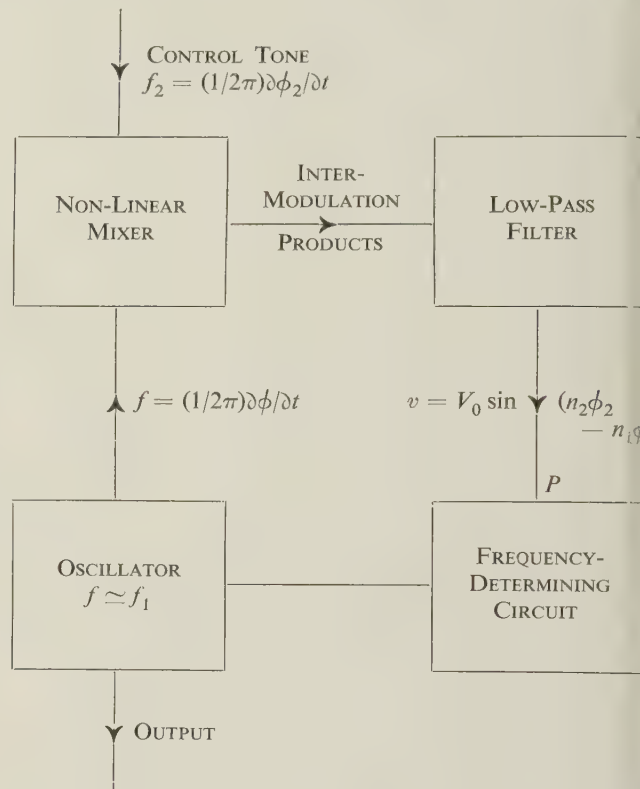


Fig. 1. Principle of frequency-control

## THEORY

Referring to Fig. 1, let the oscillator to be controlled and the controlling oscillator have instantaneous phases  $\phi$  and  $\phi_2$  such that

$$n_2\phi_2 + \text{constant} \equiv n_1\phi_1 \simeq n_1\phi \quad (1)$$

where  $n_2$  and  $n_1$  are integers,  $\phi_1$  is the desired phase of the oscillator and  $n_2/n_1$  is therefore the control ratio. The corresponding frequencies will be

$$\left. \begin{aligned} f_2 &= (1/2\pi)\partial\phi_2/\partial t \\ f_1 &= (1/2\pi)\partial\phi_1/\partial t \\ f &= (1/2\pi)\partial\phi/\partial t \\ f &\simeq f_1 \end{aligned} \right\} \quad (2)$$



Let both oscillations be applied to a non-linear mixer such that the output voltage contains beats of phase  $n_2\phi_2 - n_1\phi_1$ . Beats  $m\phi_2 - n\phi_1$ , where  $m$  and  $n$  are any pair of integers, will, in general, also be present, these may be removed by a low pass filter. Then the output voltage of the mixer,

$$v = v_0 \sin(n_2\phi_2 - n_1\phi_1) \quad (3)$$

is applied to a frequency determining device (such as a reactance valve) coupled to the oscillator.

The oscillator frequency may now be expressed by the equation:

$$f = (1/2\pi)\partial\phi/\partial t = f_1 + \Delta f + kv \quad (4)$$

where  $f_1 = (1/2\pi)\partial\phi_1/\partial t$  is the desired controlled oscillator frequency,  $\Delta f$  is the departure of the oscillator from its value due to drift and imperfect initial tuning, and  $k$  is the correction which is applied through the frequency controlling element. Combining (3) and (4) we have:

$$f = f_1 + \Delta f + kV_0 \sin(n_2\phi_2 - n_1\phi_1) \quad (5)$$

Now  $f_2$  and  $f_1$  are constant by definition. Suppose that there is a solution of equation (5) such that  $f$  is independent of time, then the only term involving time-dependent parameters is the last and this, too, must be constant.

Equations (2) may now be integrated:

$$\left. \begin{aligned} \phi_2 &= 2\pi f_2 t + \alpha \\ \phi_1 &= 2\pi f_1 t + \beta \end{aligned} \right\} \quad (6)$$

hence  $n_2\phi_2 - n_1\phi_1 = 2\pi t(n_2f_2 - n_1f_1) + \theta = \text{a constant}$

where  $\theta = n_2\alpha - n_1\beta = \text{a constant}$ .

Thus  $n_1f = n_2f_2 \equiv n_1f_1$

and, hence,  $f = f_1$ .

We also find that  $\Delta f + kV_0 \sin \theta = 0$ , which implies that the oscillator frequency is locked at the desired value of  $f_1$  and that the phase difference  $n_2\phi_2 - n_1\phi_1 = \theta$  is adjusted to compensate for  $\Delta f$ . The range of  $\Delta f$  over which locking is maintained is clearly  $\pm kV_0$ .

This simple treatment does not settle the question of how rapidly changes of  $\Delta f$  will be corrected; indeed it implies that correction will be instantaneous. Closer consideration shows that the controlled oscillator may give the correct mean frequency but may oscillate about its value at some low frequency due to the finite rate at which the oscillator responds to the correction voltage and due to the time delay introduced by the low pass filter circuit. Mathematical treatment of this problem is quite feasible but is hardly profitable since the method and result are intimately related to the properties of the particular system used. However, the general condition which must be fulfilled for stability can readily be stated as follows.

Suppose that the control loop is broken at  $P$  and that a voltage  $V + v(\omega)$  is applied to the frequency controlling element, where  $V$  is a steady component maintaining the correct mean frequency and phase, and  $v(\omega)$  is a small alternating component of a low frequency  $\omega$ . The oscillator will then be phase modulated, leading to an output voltage  $V + v'(\omega)$  from the mixer and low pass filter circuits. If the equation

$$v'(\omega) = v(\omega) \quad (7)$$

is satisfied both in magnitude and phase for any particular value of  $\omega$ , phase oscillation will result when the control loop is closed and the steady state solution of equation (5) will not be realized. The problem is, in fact, one analogous to that of feedback amplifier design in which Nyquist's theorem may appropriately be applied.

#### A PRACTICAL ARRANGEMENT

In Fig. 2, the circuit is given of a practical arrangement which has met the need expressed earlier, for a generator of stable audio frequencies. An  $R-C$  oscillator of the Wien Bridge type with lamp stabilization is used and its frequency is controlled through the reactance valve  $V_3$  which has only a relatively small effect on the frequency (a few %). The output from the oscillator is amplified by  $V_1$  to a level about 50 V r.m.s. This is applied through a limiting resistor to the control grid of  $V_2$  a 6L7 pentagrid mixer valve. Similarly, the voltage from the 1 000 c/s tuning-fork is stepped up to a level of about 50 V, limited and applied to the third grid of  $V_2$ . Thus the input waveforms of the first and third grids of  $V_2$  are both rectangular. (That portion of the negative grid swing beyond anode current cut-off is clearly ineffective.)

The anode current of  $V_2$  contains very many intermodulation products and all but the very low frequency components are attenuated by a simple low pass filter composed of the anode load resistor in parallel with a capacitance giving a time constant of 0.04 sec or a characteristic frequency of 4 c/s. This load is directly coupled to the grid of the reactance valve  $V_3$ .

The performance of this arrangement was observed by connecting the X and Y plates of a cathode ray oscilloscope at the points X and Y in Fig. 2. It was found that the  $R-C$  oscillator was locked whenever it was tuned close to a frequency  $f_1$  where  $n_1f_1 = n_2f_2$ ;  $n_1$  and  $n_2$  might have integral values as high as 30. Small externally induced fluctuations of  $f$  led to very rapid readjustment (of the order of 0.1 sec). There was no tendency to oscillation of frequency about the correct value, in the manner considered earlier, but this could be produced by introducing an additional  $R-C$  filter between the mixer and the reactance valve. This is to be expected since the oscillator will exhibit some inertia in responding to changes of control voltage and this, with a two-stage filter network, can lead to the conditions needed to satisfy equation (7).

Although locking occurred at a very large number of frequencies over a wide range (from about 100 c/s–10 kc/s), the component values were chosen to give optimum performance in a much more restricted region. The range 2.0–2.5 kc/s was examined in detail and it was found that locking occurred at the frequencies shown in the Table. Those frequencies marked with an asterisk were securely locked and could be maintained indefinitely. The remainder were locked but not sufficiently well to remain so without retuning the oscillator at intervals. The locking range  $\pm\Delta f$  was greater than 0.2% of  $f_1$  at half of the controlled frequencies and was 1.5% in the best case (at 2.0 kc/s). The harmonic distortion of the oscillator output and the amplitudes of the 1 000 c/s note and other unwanted frequencies were negligible.

However, in a few cases, particularly at the higher values of  $n_2$  and  $n_1$ , there was evidence of slight phase modulation of the controlled oscillator. This is not to be confused with the type of instability mentioned earlier. Consideration of the functioning of the mixer shows that, when locking has occurred, its anode current contains only one set of frequencies: the highest common factor  $F$  of the two frequencies  $f_1$  and  $f_2$ , viz.,

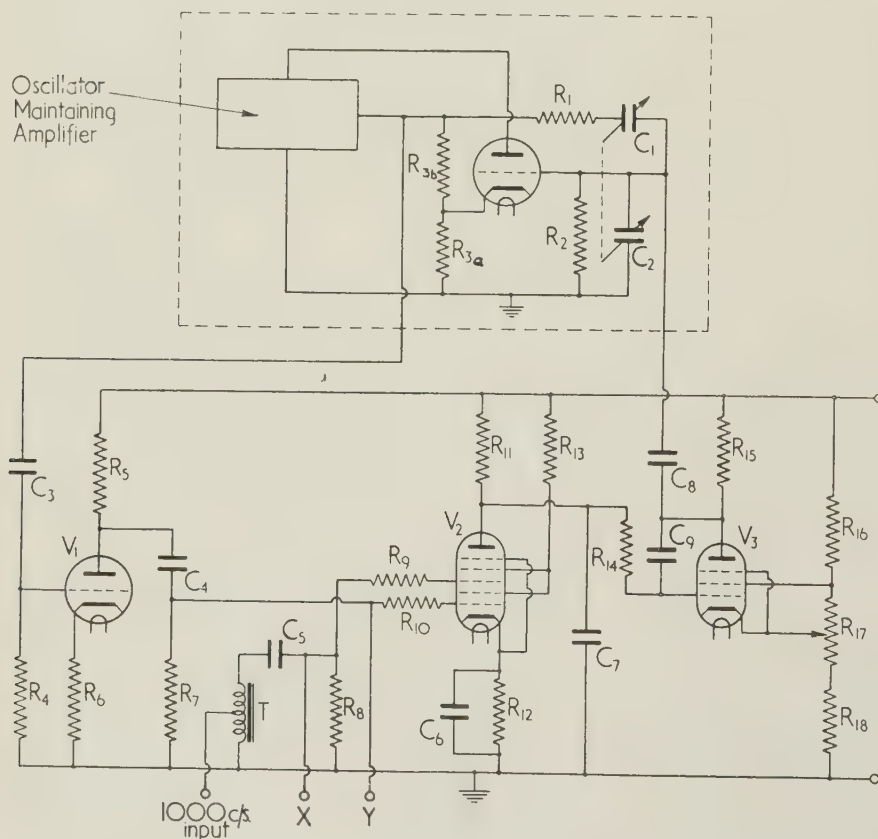
$$F = f_2/n_1 = f_1/n_2 \quad (8)$$

Frequencies at which locking occurred within 2.0–2.5 kc/s range

1 c/s	$n_2$	$n_1$	1 c/s	$n_2$	$n_1$
*2 000.0	2	1	2 300.0	23	10
2 100.0	21	10	2 307.7	30	13
2 111.1	19	9	*2 333.3	7	3
*2 125.0	17	8	2 357.1	33	14
*2 142.9	15	7	2 363.6	26	11
2 166.7	13	6	2 375.0	19	8
2 181.8	24	11	*2 400.0	12	5
*2 200.0	11	5	*2 428.6	17	7
2 222.2	20	9	2 444.4	22	9
*2 250.0	9	4	2 454.5	27	11
2 272.7	25	11	*2 500.0	5	2
*2 285.7	16	7			

\* Frequencies locked and maintained.

together with the harmonics  $2F$ ,  $3F$  . . . and a steady component. It is the latter which maintains the oscillator locked, the others being filtered out. However, when  $n_1$  and  $n_2$  are large,  $F$  is low and may not be completely eliminated in which case it might be passed on to the reactance valve. Other effects are also possible should a third frequency—for example that of the mains—be present in the mixer circuit.



Component values

$R_1 = R_2 = 76.8 \text{ k}\Omega$ (for 1.4–5.1 kc/s range)	
$R_{3a}$ = Oscillator stabilizing lamp (3rd arm of Wien Bridge)	
$R_{3b}$ = 4th arm of bridge	
$R_4$ = 500 k $\Omega$	
$R_5$ = 220 k $\Omega$	
$R_6$ = 5 k $\Omega$	
$R_7$ = 2 M $\Omega$	
$R_8$ = 2 M $\Omega$	
$R_9$ = 560 k $\Omega$	
$R_{10}$ = 560 k $\Omega$	
$R_{11}$ = 20 k $\Omega$	
$R_{12}$ = 3.3 k $\Omega$	
$R_{13}$ = 15 k $\Omega$	
$R_{14}$ = 56 k $\Omega$	
$R_{15}$ = 2 M $\Omega$	
$R_{16}$ = 10 k $\Omega$	
$R_{17}$ = 100 $\Omega$ wire wound variable	
$R_{18}$ = 22 k $\Omega$	
$C_1$ = 400–1 300 $\mu\mu\text{F}$	
$C_3$ = 750 $\mu\mu\text{F}$	
$C_4$ = 0.1 $\mu\text{F}$	
$C_5$ = 0.1 $\mu\text{F}$	
$C_6$ = 50 $\mu\text{F}$	
$C_7$ = 2 $\mu\text{F}$	
$C_8$ = 150 $\mu\mu\text{F}$	
$C_9$ = 30 $\mu\mu\text{F}$	
$V_1$ = 6J5	
$V_2$ = 6L7	
$V_3$ = EF39	
$T$ = 2/1 transformer	

Fig. 2. A practical circuit for frequency controlling an R.C. oscillator



## CONCLUSION

A method of controlling an oscillator at a multiple or sub-multiple of the controlling frequency, or in a  $\pi$ -integral rational ratio, has been described with particular reference to the provision of a discrete set of highly stable audio frequencies. The practical arrangement is capable of further development by separation of the different functions of the mixer valve. For example, a sine-wave controlling tone and the output of the oscillator might be used to produce two synchronized trains of pulses operating a "gate" circuit such that only coincident or partially coincident pulses would contribute to the output current. The d.c. component of the latter could then be used to effect control of the oscillator circuit.

Although the work described has been concerned exclusively with audio oscillators, the principle is clearly

a more general one, and it is possible that u.h.f. or even microwave oscillators could be frequency-controlled in a similar manner.

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## SPECIAL REPORT

## Report of Discussion on Calibration Line Drift in Spectrographic Analysis—Sheffield, February 1950

The Industrial Spectroscopy Group of The Institute of Physics and the Sheffield Spectrographic Discussion Group held a joint meeting in the University of Sheffield on 17 February, 1950, to discuss calibration line drift in spectrochemical analysis. The papers and the discussion on them are summarized in this report.

The meeting was opened by Mr. H. R. Clayton (British Aluminium Co. Ltd.), who summarized a number of contributions. Short papers were presented by: Mr. H. R. CLAYTON, Mr. R. J. WEBB (Ministry of Supply, Woolwich), Mr. R. DIXON (William Jessop and Sons Ltd., Sheffield), Dr. R. O. SCOTT (Macaulay Institute, Aberdeen) and Mr. W. E. VAN R. DE JONG (N.V. Hollandse, Metallurgische Bedrijven).

Mr. Dixon presented results obtained by the Sheffield Spectrographic Discussion Group: and the contribution made by Mr. Clayton summarized work in his own laboratories as well as the laboratories of Dr. Addink (N.V. Philips Gloeilampenfabrieken, Eindhoven), Mr. Munwoody (Northern Aluminium Co. Ltd.), Mr. S. Lipling (Wolsey Motors Ltd.), Mr. E. van Someren (Murex Welding Processes Ltd.), and the Post Office Engineering Department.

The short papers dealt with different aspects of calibration drift and covered a variety of types of material and excitation. The discussion which followed was very

active and as it dealt with many points not covered in the introductory papers the following brief account summarizes both the papers and the discussion irrespective of the source of the information.

## DRIFT ARISING IN THE DISCHARGE

Evidence was produced showing that with d.c. arc excitation of powdered mineral samples in craters in graphite electrodes, variations in the depth or diameter of the crater influence the index point of the calibration graph and that the tightness with which the powder is packed into the crater is also of great importance. The effect of background variations in this type of analysis was also stressed, and graphs were presented to show the advantages of background compensation, particularly when wide ranges of concentrations (300-fold) are to be covered. Under these conditions curves uncorrected for background show both lateral and angular shift whilst those to which a background correction is

applied show only lateral shift, which can be minimized by careful electrode preparation and packing.

Index point shift may be artificially produced in circuits of the Pfeilsticker type by altering the relative powers of the high and low voltage sections of the discharge and in the analysis of aluminium, silicon shows this effect to a marked degree. It was suggested that the shift produced by these changes might occur in practice because of uncontrolled variations in gap breakdown potential. It was shown that with circuits of this type the variations in mains voltage had no significant effects on index-point drift, and only affected the overall intensity of the spectrum.

In steel analysis it was reported that the extent of index-point drift was unaffected by temperature, humidity, surface finish of the electrodes, or spectrograph and microphotometer slit width, whilst workers with aluminium noted that drift could not be correlated with temperature, pressure, operator or time but showed a significant correlation with mean plate density. One laboratory reported a seasonal trend on the index point which appeared to rise with the relative humidity from spring to summer, but an attempt to confirm this by operating the discharge in a saturated atmosphere gave contradictory results. Introduction of steam around the discharge caused a shift of index point for manganese and copper in steel, but not for other elements. From figures supplied by numerous contributors it was established that in non-ferrous analysis the variation of index point amounted to  $\pm 6\%$  of the mean, whilst the slope of the calibration curve had a standard error of some 8% of the mean value.

In steel analysis, using a condensed spark circuit with no added inductance, small variations ( $5\ \Omega$ ) in circuit resistance had no effect on the index point for vanadium but changes in the shape and material of the counter-electrode, used in conjunction with a flat test sample, produced a shift.

#### DRIFT DUE TO PHOTOGRAPHIC RECORDING

Records from two laboratories showed that if satisfactory methods of plate calibration are used changes in index point due to variations in emulsion characteristics are negligible. It was noted by one investigator that the relationship between contrast factor and wavelength for one particular type of plate had shown variations over a period of years. Two laboratories had made

systematic studies of the effect of humidity on the sensitivity of the photographic plate and changes were observed when the relative humidity was altered from 36% to 46% in one case, and from 50% to 85% in another. The variations were most marked at the longer wavelengths and this was considered to be due to the change in the light absorption of the silver bromide of the emulsion with wavelength.

The contribution of the photographic recording and measurement to the total drift calibration had been estimated in an experiment in which two spectrographs were used to produce simultaneous records of a single source. Statistical examination of the results which covered 12 plates and 36 estimates of the calibration parameters, showed that the drifts in index point and in slope were due partly to the photographic side and partly to the electrodes and their excitation, the distribution being approximately evenly divided in each case.

#### ERRORS IN MICROPHOTOMETRY

More than one speaker emphasized that any microphotometer employing a finely focused light beam for exploring the spectrum line depends for its precision on accurate focusing, which may in turn depend on the uniformity of thickness of the photographic emulsion and of the glass plate itself. The stability of the instrument adjustments and the vigilance of the operator are also important factors. It was noted by two investigators that the focus setting obtained by the parallax method is not necessarily the optimum and for most accurate work a microphotometer should be focused by trial and error so that the apparent density of a line is a maximum. Research on the microphotometry of spectra on the same plate by different laboratories showed that different observers and instruments not only obtained different readings for the same lines, but also different ratios of line intensities. The critical parameter was considered to be the ratio of microphotometer/spectrograph slit width, and the importance of this factor varied with different spectra according to the relative sharpness of the lines used. It was agreed that insufficient attention was paid to microphotometer design and in particular that information of value could be obtained by the use of a scanning microphotometer to examine the density profile of the lines used.

H. R. CLAYTON, E. VAN SOMEREN



## NOTES AND NEWS

## New Books

**rials for Centimetre Wave-lengths.** By D. W. FRY and F. K. GOWARD. (London: Cambridge University Press.) Pp. vi + 172. Price 18s. net.

Many engineers and inventors do not appreciate the limitations of aerial performance, and tend to embark on centimetre wave projects without realizing the difficulties of meeting their requirements in regard to polar diagrams and impedances. They should read this book, which devotes a great amount of its space to describing what has been done in the field of centimetric radar to achieve specified polar diagrams. This is demonstrated, not by data curves and tables, but by a critical discussion of the pros and cons of many alternative design methods.

The discussion always mentions manufacturing tolerances where these are of importance, and is illustrated by sketches giving typical measurements, intended as a design guide, but not as manufacturing information.

The technical reader is assumed to possess sufficient mathematical and experimental technique to appreciate the points made, and to follow the advice of the authors. It is therefore a valuable source book to the serious designer of aeriels who is not entirely inexperienced. The bibliography has been carefully compiled to supplement the information given in the book and includes references to the majority of recent publications in the field.

W. A. JOHNSON

**Survey of General and Applied Rheology.** By G. W. SCOTT BLAIR. (London: Sir Isaac Pitman and Sons, Ltd.) Pp. xv + 314. Price 40s. net.

The treatment of the subject is that of rheology and the book can be of value to workers with problems of consistency. But many may regard it as more useful as a bibliography than as a text book giving fundamental theoretical approaches which could be applied to any system. Admittedly this is difficult and no such complete theoretical approach exists. However, the beginnings have been made but the reader will not learn much from this book. It is to Scott Blair's credit that he has mentioned such a large number of original papers, many not easily accessible, and that he has attempted to make more of a bibliography. But no attempt seems to have been made to emphasize papers of basic and fundamental importance. The way in which the subject matter is divided into chapters seems to indicate lack of clear planning, but the basic confusing cause is the author's view point. The danger of following a highly specialized subject especially when it is crisscrossed with an esoteric name is that it leads to an esoteric outlook. Rheology will never become a science until it ceases to try to be rheology.

The distinction is made quite early in the book that enquiries into Rheology fall into the analytical school which regards a material "about equally removed from elastic and viscous behaviour" as consisting of two parts, one viscous and the other elastic, and the effects of these parts are considered as additive. . . . The second school seeks a simple and initially empirical relation between stress, strain and time, and only rarely accepts the revolutionary change of quantity into quality. The wording is typical of the whole

book: the description of the analytical school is inept and gives no clear idea of its scope and achievement; that of the second is clear and accurate, while the final phrase is an example of an almost mystical philosophical allusiveness which keeps intruding.

After a historical introductory chapter, Chapters II and III entitled "Ideal Materials" deal with Hookean and Newtonian systems. Chapter III, however, passes rapidly from the Rate Process theory of fluid flow to high-polymer solutions ending with a reference to a Rotatory Reynolds number without the faintest clue to the hydrodynamical theory which postulates the precession motion which could reach a Critical Rotatory Reynolds value. The Einstein equation is given, as usual, without mention of its hydrodynamical derivation consequently the logical development of the results of anisometry is lost. Indeed, there is no clear differentiation between Newtonian and non-Newtonian systems in this chapter on ideal liquid systems.

Chapter IV discusses the formal classification of deformation of systems intermediate between liquids and solids and then suddenly passes to the rheology of cervical secretions and bovine pregnancy. Chapter V deals with thixotropy and other cases of fall of consistency with shearing. On the first page Scott Blair discusses "ordered structure" and explains in a footnote that "structure" has here the sense of the German "Struktur" but the offending word is "ordered."

Reduction of consistency by shearing is discussed, but the discussion of thixotropy leads nowhere. Langmuir's work showing that thixotropic recovery is a Rotational Brownian disorientation is not mentioned, although it is badly misquoted in Chapter VII; nor in this section, where it belongs, is the work of J. R. Robinson, Boeder and others who have explained so clearly the nature of pure hydrodynamical anomaly in systems of non-spherical particles. Goodeve's theory of thixotropy is badly described insofar as the Rate Process idea is not mentioned—namely that the apparent consistency is a balance between a rate of breaking (by shear) and rate of remaking of bonds. The criticism that this is an explanation of "structural viscosity" rather than of thixotropy is sound.

Chapter VI deals with increase of consistency caused by shearing but contains little of value. Chapter VII, after a brief paragraph on orientation of charged colloid particles, passes to a most unsatisfactory description of liquid crystals. Orientation of rod-shaped particles by flow is qualified by the statement: "This orientation is presumably statistical since the particles must be rotating under the influence of the viscous couple." The chapter ends with stickiness and oiliness.

Chapter VIII discusses rheological measurements and contains an attempted formal classification based, however, on rather informal properties. It ends with an exhaustive list of references classified first under the type of measuring apparatus with subclassification into the types of material examined. Chapters IX, X and XI discuss types of viscometers. Part II of the book is entitled "Rheological Interpretations and the evidence of psychophysical investigations." Chapters XII and XIII—"the analytical school"—starts with

Maxwell's relaxation theory and spring and dashpot models, while Chapter XIV—"the integrative school and other treatments"—deals with power laws and the Nutting equation. The reviewer finds little difference between these two alleged schools. The final 35 pages of the book embody psychophysics and gestalt psychology, which the reviewer considers unhelpful in this book.

There are in effect three Indexes: first "Summaries of Rheological papers on special subjects" in which the summaries are so brief that its value is reduced. This is followed by a bibliography, classified under authors' names and finally the orthodox author and subject indexes.

The book is called a Survey: any author has the right to make his survey from any view point that he thinks fit, but he also accepts the duty of justifying this view point. I consider that Scott Blair does not do this because his psychological leading strings muddle the subject rather than clarifying it for the reader. There are also far too many intermissions on a personal note for a scientific book.

A. S. C. LAWRENCE

## Journal of Scientific Instruments

### Contents of the June Issue

#### *The 34th Physical Society's Exhibition*

The Craftsmanship and Draughtsmanship Competition.  
By H. W. B. Gardiner.

Electronic Instruments. By A. J. Maddock.

Non-Electronic Electrical Instruments. By A. Felton.

Optical, Mechanical, Thermal and Vacuum Instruments.  
By H. Barrell.

A High-Temperature Debye-Sherrer X-ray Camera. By  
E. C. Williams.

A Portable Battery-Operated Radiation Monitor. By F. B.  
Whiting, D. G. A. Thomas and J. B. Marsh.

A Calorimeter for the Measurement of Small Amounts of  
Heat liberated slowly. By W. Hirst and M. Kerridge.

Accurate Adjustment of the Wedges of a Soleil Compensator.  
By H. G. Jerrard.

A Simply Constructed Vortex Tube. By M. P. Blaber.

A Direct-Reading Instrument for the Measurement of the  
Series-Resistance of Capacitors. By F. Gutmann.

A Machine for rapidly processing Photographic Trace Re-  
cordings. By G. I. P. Levenson.

A Sensitive Adjustment for Interferometer Plates. By O. S.  
Heavens.

### NOTES AND NEWS

#### *New Instruments, Materials and Tools*

Valve Voltmeter—Electronic Flash Tubes—Resistance-Capa-  
city Oscillator and Frequency Monitor—Sub-Miniature  
Valves for Hearing Aids—35 mm Film Attachment for  
Quartz Spectrographs—Thin Copper Foil—Co-axial  
Air-spaced Cables—Microsecond Counter Chrono-  
meter—Lens Testing Charts.

#### *New Books*

Counting Tubes—Glass-to-Metal Seals—Radio Servicing  
Equipment—An Introduction to the Theory and Design  
of Electric Wave Filters—Cathode-ray Tube Traces—  
Flow Measurement and Meters—The 24 m-Interference  
Comparator of the Geodetic Institute at Potsdam—  
Practical Spectroscopy.

**Spectrophotometry.** By KASSON S. GIBSON. (Washing-  
ton: U.S. Government Printing Office.) Pp. 44.  
Price 25c.

This booklet is one of the series of Circulars issued by the  
National Bureau of Standards, Washington, its primary  
purpose being to acquaint users of spectrophotometers with  
the experience, methods and data resulting from the Bureau  
work on this subject. Notes are also included on available  
standards of spectral emission, transmission and reflexion.

With the exception that infra-red spectroscopy, viz. radi-  
metric methods applied at wavelengths greater than  $1\mu$ ,  
deliberately excluded, Gibson gives in a short space  
remarkably complete authoritative account of the principles  
and instrumental methods in general use for both physical  
and chemical investigations. The chapter on errors and their  
elimination is of particular importance and should be noted  
carefully by all workers new to the field. An excellent  
bibliography on the practice of spectrophotometry, but no  
applications, is included.

B. S. COOPER

**Temperatures in a Test Bungalow with some Radiant  
and Jacketed Space Heaters.** (Washington: U.S.  
Government Printing Office.) Pp. 44. Price 33c,  
post paid. This is NBS Building Materials and  
Structures Report BMS 114.

## British Journal of Applied Physics

Special Articles and Original Contributions accepted for publica-  
tion in future issues of this Journal

### SPECIAL ARTICLES AND REPORTS

Electrical Conduction in Glass. By P. L. Kirby.

Summarised Proceedings of Conference on X-ray Analysis—  
Leamington Spa, 1949. By A. E. De Barr and C. H.  
Carlisle.

### ORIGINAL CONTRIBUTIONS

The Rheology of Unseasoned Linoleum. The Significance  
of Fillers in Calendering. By F. T. Walker and J. Arnold.

A Method for Measuring the Efficiency of Getters at Low  
Pressure. By S. Wagener.

Mechanical Models for the Representation of Visco-Elastic  
Properties. By R. Roscoe.

Improvement of the Field of a Solenoid by End Coils. By  
B. Lewis.

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## SPECIAL REPORT

Summarized Proceedings of a Conference on X-ray Analysis—  
Leamington Spa, 1949

A Conference of the X-ray Analysis Group of The Institute of Physics was held at the British Iron and Steel Corporation Ltd.'s House, "Ashorne Hill," near Leamington Spa, on 2nd and 3rd December, 1949. The first session was devoted to a discussion on "The interaction of structure and chemical behaviour" and the second to "Geiger counter techniques in X-ray analysis." The papers and the discussion on them are summarized in this report. Reference is also made to Professor C. A. Coulson's lecture on "Chemical bonds, their shape and their behaviour," and a brief summary of Professor M. von Laue's address on "The absorption of X-rays" is included.

THE INTERACTION OF STRUCTURE AND  
CHEMICAL BEHAVIOUR*Precision X-ray Analysis and Crystal Structure  
Determinations.*

R. D. W. J. CRUICKSHANK (University of Leeds) read the first paper of the opening session on "Some recent theoretical developments in the precision X-ray analysis of molecular structure."<sup>(1)</sup> He pointed out that atomic co-ordinates derived from uncorrected electron density maps are liable to certain errors, some systematic and some random. The two sources of systematic error are the use of a finite Fourier series of structure factors and the thermal motion. Owing to these causes the maximum of one peak is displaced by diffraction ripples and the spreading out of other peaks. These systematic errors can be corrected by Booth's<sup>(2)</sup> method of reversing the displacements found in syntheses of calculated structure factors based on the uncorrected atomic co-ordinates.

Random errors occur because of experimental errors in intensity measurement, computational approximations and errors in the unit cell size. Some allowance must also be made for the imperfection of the correction of systematic errors. If the computational and cell errors are small (as is usual), a good estimate of the standard deviation of a peak co-ordinate due to random errors is in orthorhombic cells)

$$\sigma(x) = \frac{1}{V} \frac{2\pi}{a} \left( \sum_3 h^2 \Delta F^2 \right)^{\frac{1}{2}} \left/ \left| \frac{\partial^2 \rho}{\partial x^2} \right| \right. \quad (1)$$

where  $\Delta F = |F_{\text{obs}} - F_{\text{calc}}|$  and  $\partial^2 \rho / \partial x^2$  is the second derivative at the maximum.

In many structure problems of current interest the effects sought after are of much the same order of magnitude as the errors. In these cases it is essential for the full and valid interpretation of the results to use statistical significance tests. It seems unlikely that any startling improvement in accuracy with the same experimental data can be achieved either by weighted Fourier series with corrections for systematic errors or by the least squares method; the two methods having been shown by Cochran<sup>(3)</sup> to give the same atomic co-ordinates.

DR. C. J. BROWN (Imperial Chemical Industries Ltd.) followed with "The crystal structures of an isostructural

group of organic compounds."<sup>(4)</sup> The crystal structure of aniline hydrochloride had recently been determined in their laboratories. The molecules are arranged in the unit cell which has dimensions  $a = 15.84 \text{ \AA}$ ,  $b = 5.33 \text{ \AA}$ ,  $c = 8.58 \text{ \AA}$ , and  $\beta = 101^\circ$ . Only two-dimensional Fourier refinement was undertaken as the crystals obtainable showed marked cleavage and had a moderately high absorption, so it was thought that intensity data may not be reliable enough to warrant continuing the work. The benzene ring is apparently regular, but the C-N bond had an apparent bond order of about 1.5, in common with many other C-N bonds in a wide variety of organic compounds. The chlorine ions and nitrogen atoms have three nearest neighbours.

Shortly after the completion of this structure determination, a problem concerning *p*-aminophenol was submitted and it was noticed that two of the cell dimensions bore a resemblance to those of aniline hydrochloride. The suggestion was made that the molecular packing in these two directions would be the same, and this has indeed proved to be the case. The threefold ionic co-ordination in the one case is replaced by three hydrogen bonds in the other. The structure has been completely refined by three-dimensional syntheses and the hydrogen bond lengths are 2.88, 3.11 and 3.13 Å. It may be that the shortest is due to the -OH hydrogen while the others are from the -NH<sub>2</sub> hydrogens. The C-N bond apparently does not shorten in this case, and has a bond order of 1.0.

As a result of a search of the literature it was found that a large number of aromatic organic compounds crystallize so that two of their cell dimensions are roughly 5.5 and 8.5 Å, owing to the packing of their benzene rings. The angle between the benzene rings is always about 60°, presumably because of their elliptical cross-section, which gives this angle as the most economical for close packing.

One use of this classification into an isostructural group is in the case of polyphenylene and poly-*p*-zylylene, powder photographs of which had recently become available. Although there were only about a dozen lines on the photographs, it was possible to assign a unit cell on the basis of analogy with this group of compounds, and from the indices of the rings, a

good general picture of the arrangement of the molecules in the polymer was found.

There is evidence that many other paradisubstituted and monosubstituted aromatic compounds crystallize with this two-dimensional packing, and it is possible that ortho- and meta-substituted benzene compounds form isostructural systems of their own, but further classification of this kind is a problem for the future.

The third paper of the afternoon was read by MR. E. J. W. WHITTAKER (Ferodo Ltd.) on "Some recent work on the structure of fibrous silicates."<sup>(5)</sup> He said that in view of the relatively long time which has elapsed since any structural work was done on an amphibole mineral, the time is ripe for a redetermination of the amphibole structure. This has now been done for Bolivian crocidolite, which is particularly suitable both for theoretical and practical reasons; theoretically because no type of crocidolite has previously had its structure determined, and practically because it occurs in the form of fibres which approximate to single crystals. The structure that has been found agrees in outline with that deduced for tremolite by Warren,<sup>(6)</sup> but a number of finer details emerge. It is found that the silicate chain is not planar but curved so that it lies a little farther from the plane of the metal ions at the edge than at the centre. This is explained on the same lines as those which led to the postulation of curved layers in chrysotile.<sup>(7)</sup> Other minor distortions from the regular arrangement given by Warren are also believed to occur. The distribution of the metal ions among the available non-equivalent sites is found to be non-uniform, and the observed distribution can be qualitatively explained by the effect on the charge distribution produced by the different propensities of the ions present for six-fold and eight-fold co-ordination. Further problems connected with isomorphous substitution in the amphiboles were pointed out, and a possible way was indicated of accommodating extra water molecules in holes in the structure, in excess of the normal amount of combined water.

In more general work on fibrous silicates evidence of disordered amphibole structures has been found in anthophyllite from Finland and in the monoclinic amphiboles amosite and montasite. In the case of montasite, photometric measurements of fibre photographs taken with an elongated slit and crystal reflected radiation have demonstrated that the disorder is connected with longitudinal displacements of the chains. New evidence has been found of variations in structure of various specimens of chrysotile from different sources which give fibre photographs differing considerably on the second layer line, and three types were illustrated. A fibre photograph of paligorskite has been obtained with fairly well developed orientation, and it has been shown qualitatively that this mineral possesses the same structure as was found by Bradley for attapulgite.<sup>(8)</sup> Such a fibre photograph provides more extensive diffraction data than was previously available from the type of oriented aggregate which could be prepared with attapulgite.

### *X-ray Analysis and Infra-red Spectra.*

The last paper, "The structure of the cyanoethyl celluloses," was presented jointly by Dr. F. HAPPEY, DR. J. H. MACGREGOR and MR. I. F. TROTTER (Courtaulds Ltd.)<sup>(9)</sup> and read by Dr. Happey.

The cyanoethyl celluloses can be roughly classified into three types: (a) alkali soluble, water insoluble; (b) water soluble; (c) acetone soluble.

There is no direct correlation between the solubility and the degree of substitution and it is possible to have two cyanoethers of the same degree of substitution which have different solubility properties. In class (a) the degree of substitution covers the range 0.15–1.70 groups per glucose residue, in (b) from 0.6–1.2, and in (c) 2.5–3.0. Between the limits 1.7–2.7 it is necessary to use mixed solvents, but in no case can the limits be strictly defined.

In general, it has been found possible to distinguish the three types of material by X-ray methods. The X-ray diffraction photograph of fibres of the alkali soluble material of low substitution is somewhat similar to that of hydrate cellulose and falls off in definition with increased degree of substitution due to distortion of the crystalline lattice caused by replacement of hydroxyl by cyanoethyl groups. The X-ray diagram of the water soluble derivatives is almost amorphous in character, while that of the alkali soluble derivatives of higher substitution shows some increase in crystallinity with increase in substitution of the cellulose. Fibres can be spun and crystallized from acetone solutions of the cyanoethers of class (c). The structure so formed presents some difficulties in interpretation as it is not fully substituted, but the X-ray diagram can be indexed to a first approximation on a hexagonal lattice with a unit cell as follows:

$$\begin{aligned} a &= c = 9.74 \text{ \AA} \\ b &= 15.21 \text{ \AA fibre axis} \quad \beta = 120^\circ \end{aligned}$$

This result would be consistent with one chain per unit cell containing three consecutive tricyanoethyl glucose residues arranged on a three-fold spiral along the fibre axis. It is, however, emphasized that this postulated structure is only provisional and may well be a sub-multiple of a larger cell.

From infra-red studies of the cyanoethyl celluloses the following correlation between the OH stretching frequency and the solubility has been established:

$\nu_{\text{OH}}$	Solubility
3 400–3 425 $\text{cm}^{-1}$	alkali soluble
3 440–3 450 $\text{cm}^{-1}$	water soluble
3 500–3 520 $\text{cm}^{-1}$	acetone soluble

These differences in the OH stretching frequency, which are indicative of different hydrogen bonding conditions, must be due to differences in the uniformity of substitution along the chains, and it has been found possible to use the OH-frequency in connexion with the known degree of substitution as a criterion of uniformity.



substitution. In the region  $950\text{--}1\,65\text{ cm}^{-1}$ , the infra-red spectra show differences which can be correlated with the three types of structure. The absorption bands in this region contain contributions from vibrations involving the single bound oxygen atoms and so would be expected to be affected by changes in the cyanoethers. At low substitution, some of the cyano groups had suffered hydrolysis to amide groups.

The general variation in the structure of the cyanoethyl celluloses with degree of substitution can be followed by x-ray diagrams of the specimens and the qualitative implications are confirmed by the infra-red absorption spectra, which also serve to amplify certain details of structure.

#### DISCUSSION ON THE ABOVE PAPERS

The discussion was opened by DR. C. H. CARLISLE (Birkbeck College, London), who noted that Mr. Cruikshank's remarks on the derivation of accurate coordinates from crystal structure determination were fully in view of the immense strides that had been made in computational methods. Carlisle, however, did stress the need at the same time for more careful experimental techniques in the accurate determination of structures, and the evaluation of results by statistical analyses was a real substitute for questionable accuracy in the course of the experimental procedure.

PROFESSOR C. A. COULSON (King's College, London) said how extremely valuable were the highly accurate bond length measurements now being made for condensed hydrocarbon molecules. In the first place they showed unambiguously for the first time that in molecules of the type of naphthalene there were definite though small disparities in the different C-C bond lengths. In the second place, an accuracy of about  $0\cdot005\text{ \AA}$  permitted a test to be made of two or three theories of the electronic structure of the molecules. Until such accuracies have been obtained in the experiments, such a test is not properly justified. It seems likely that within a few years we shall be able to use these experimental measurements to compare the degrees of validity of the various theories. For this purpose it would be particularly valuable to have about half a dozen typical molecules studied with the greatest possible precision. This is more important, for theoretical purposes, than a larger number of structures determined with less accuracy. It is a pity that this most desirable work is so relatively unexciting.

PROFESSOR J. D. BERNAL (Birkbeck College, London) remarked that in relation to Mr. Cruikshank's paper, the time had passed when a structure analysis should be undertaken for trivial reasons—that is merely to show that it could be done. Even with machines a great effort would be required for precision analysis and this should be reserved for structures where significant chemical information could be expected. There was as yet a much wider field of investigation, especially for natural substances of unknown chemical formulae, Vol. 1, JULY 1950.

where a less thorough investigation would suffice. The decision as to which substances needed to be investigated by more or less detailed analysis was bound to occupy increasingly the attention of X-ray crystallographers.

He congratulated Dr. Brown on the use to which he had put the classification of compounds on crystallographic grounds. He foresaw that this kind of classification would soon have an important part to play not only in codifying crystal structure data but in analysis itself. He pointed out that already in three fields such rational classification existed based on the possession of a common molecular cross-section in long chain aliphatic compounds, in linear polycyclic aromatic compounds, and in the sterols.<sup>(10)</sup>

In discussing Mr. Whittaker's paper PROFESSOR BERNAL said that it was indeed a pleasure to see such remarkable progress made in a field that was apparently considered to be thoroughly investigated. In his laboratory extensive investigations were now being conducted on hydrated calcium silicates and work such as Whittaker's was therefore of considerable importance. One of the compounds now being investigated was hildebrandite, an interesting fibrous silicate with a cell height of  $3\cdot6\text{ \AA}$ . This implied the existence of silica tetrahedra with their oxygen atoms actually touching. They could only be linked by hydroxyl bonds and pointed to the existence of ions  $\text{SiO}_2(\text{OH})_2$ .

DR. V. VAND (Lever Bros. and Unilever Ltd.) stated, in respect of Mr. Cruikshank's paper, that it is important to find a correct method of weighting observed structure factors for use in the least squares methods. Following on Professor Bernal's comments on Dr. Brown's paper, Dr. Vand added that it was now possible to develop a satisfactory system of classification of long chain compounds by treating the chains as uniformly scattering cylinders. Five lattice types can be recognized: *P* parallelogram (primitive), *R* rectangular (primitive), *D* diamond (or rectangular face-centred), *E* equilateral triangular (or hexagonal), *S* square. There are corresponding powder photographs to these systems. In addition finer details of classification could also take in the tilt of the chains, both with respect to the cell axes and to themselves. Dr. Vand hoped that this system<sup>(11)</sup> could be included in tables of classified structure with great advantage to the users.

MRS. D. M. HODGKIN (University Museum, Oxford) reported that yet another aromatic type had been discovered having a very short repeat unit of  $3\cdot7\text{ \AA}$ .

DR. W. COCHRAN (Cavendish Laboratory, Cambridge), speaking on Mr. Cruikshank's paper, said that the formula given by Cruikshank for the standard deviation in the electron density is equivalent to

$$\sigma(\rho) = \left[ \frac{1}{V} \int_V (\rho_0 - \rho_c)^2 dv \right]^{\frac{1}{2}} \quad (2)$$

where  $\rho_c$  is the calculated electron density and  $V$  the unit cell volume. It may be shown that the probable error in the measured electron density at a particular

point is practically independent of the magnitude of the electron density at that point. These results suggest that the standard deviation may be taken simply as

$$\sigma(\rho) = (\overline{\rho_0^2})^{\frac{1}{2}} \quad (3)$$

where the average is taken over a number of points where one would expect the true value of the electron density to be zero. Analogous results for the standard deviation of an atomic co-ordinate, or the standard deviation remaining after correction of this result for the termination of the Fourier series, can be obtained.

MR. CRUIKSHANK replied that it was possible to estimate in advance how many planes would be needed to determine a structure with a given accuracy. For a given structure

$$\sigma(x) \propto 1/\sum_3 \omega h^2 f^2 \quad (4)$$

$\omega$  being the weight assigned to a plane and  $f$  the scattering factor. He agreed with Dr. Vand about the difficulty of weighting planes at the start of a refinement, but was uncertain whether it was very important.

PROFESSOR J. M. ROBERTSON (University of Glasgow), in referring to Mr. Cruikshank's paper, stated that in the recent three dimensional work on naphthalene<sup>(12)</sup> no mathematical corrections of the kind described had yet been applied to these results, but felt that such corrections, owing to the rapid convergence of the series, would have only a small effect on the final values of the parameters. In three-dimensional work generally it was perhaps even more important to make a large number of sections and carefully to explore the exact shape of the electron distribution. Misleading results could easily be obtained if only one or two sections at arbitrary levels were drawn through each atom. If the electron distribution was not strictly spherical, on account of preferred thermal movements, then the arbitrarily drawn section might give a false impression of the position of the mean centre of the atom.

With reference to Dr. Brown's paper, PROFESSOR ROBERTSON drew attention to another series of related organic structures which comprised many very large, flat molecules. Examples were the phthalocyanine structures, coronene  $C_{24}H_{12}$ , and ovaline  $C_{32}H_{14}$ . In all these monoclinic crystals the  $b$ -axis had a length of about 4.7 Å and the molecular plane was inclined to (010) at an angle of about 45°. The perpendicular distance between the planes of overlying molecules was very close to the graphite interplanar distance of 3.4 Å. The planes of neighbouring stacks of oppositely inclined molecules were in all cases almost exactly perpendicular to each other.

With regard to classification of organic structures in general, and in relation to Professor Bernal's remarks, while agreeing that such crystallographic classifications were of interest and importance, PROFESSOR ROBERTSON felt that in preparing, for example, a dictionary of structures it would be difficult to get away from the more fundamental chemical classification of the type

of molecule involved. Purely crystallographic classification would often bring into juxtaposition types of molecules which were chemically incongruous. While such a classification might have its advantages, it would lead to certain difficulties in a work of reference.

MR. J. W. JEFFERY (Birkbeck College, London) asked how many of the silicate structures had been done with any accuracy to verify the tetrahedral form of  $SiO_4$ . Have any structures been worked out in which OH groups have been found attached to  $SiO_4$ ? What distortion of the  $SiO_4$  tetrahedra had been found in Whittaker's structures?

DR. HELEN MEGAW (Cavendish Laboratory, Cambridge) answered this by saying that while some of the information about  $SiO_4$  in the literature is based on *a priori* assumptions, there are exact determinations, e.g. the work of W. H. Taylor on the feldspars and its subsequent refinement by Cole and others.<sup>(13)</sup> DR. MEGAW then went on to quote the recent results of her work with reference to Professor Bernal's statement on hydrated calcium silicates, in the light of her own findings on afwillite. Up till now there had been no proved cases in any published work, so far as she knew, of the occurrence of OH directly linked to Si; in all the silicates containing OH whose structures had been worked out in detail the OH groups are co-ordinated around the other cations. Afwillite<sup>(14)</sup> turns out to contain  $SiO_2(OH)_2$  groups, the O's forming shared edges with Ca polyhedra. There is undoubtedly a system of hydrogen bonds present, but the details cannot be stated as yet until the structure has undergone further refinement to get exact values of the interatomic distances. This may open up another line of explanation for the excess water in the amphiboles referred to by Mr. Whittaker. Instead of—or as well as—occurring as "trapped" water in the hole otherwise occupiable by a cation, what appears in the analysis as water may really be present as OH groups linked directly to Si atoms.

MR. WHITTAKER, replying to Mr. Jeffery, said that his structure determination of crocidolite revealed silicon valency angles from 99° to 123°, and he agreed that regularity of the  $SiO_4$  tetrahedra had been assumed in the early work on silicate structure.

DR. I. MACARTHUR (University of Leeds), in a written communication, said that Dr. Brown's correlation of molecular orientation with quasi-elliptical axis-ratios is a local aid to structure determinations, but is scarcely novel (Müller, Bernal, Nowacki), general, nor really basic. While shape dominates, factors such as polarity, rigidity, tolerance-range and temperature response are important. Surely any fundamental classification of organic crystals must rest on potential fields and their minima? Unfortunately, for the computing theorist too, *guide* as well as *check*, precision is essential, especially in longer-range force treatment. If a descriptive basis is chosen, it should *not* be referred to three strong reflexions, say, which are indiscriminatory for long aliphatic chain compounds: second order thermodynamic properties such



anisotropic thermal expansion are here more finely indicative.

#### PUBLICATION OF PROGRAMMES OF STRUCTURE DETERMINATIONS

MR. A. ADDAMIANO (University Museum, Oxford) stated the view that it would be to the advantage of people working on structure determinations to be made acquainted with structures being investigated in other laboratories. He stressed that notification should be given as soon as the refinement of the structure was about to take place in order to prevent others from wasting their time. This should possibly be done through *Acta Crystallographica*.

DR. CARLISLE said that the suggestion made by Mr. Addamiano was worth consideration, for he had, himself, found that work about to be taken up by him had been started in other laboratories. He felt that laboratories could co-operate to the general good by arranging to have half-yearly or yearly programmes of their work published together with intended programmes.

PROFESSOR ROBERTSON felt, however, there might be serious difficulties on publication of intended structure determinations by different laboratories, chiefly for the reason that the very best of intentions were so frequently not fulfilled. He had many structures of his own that he was trying to solve and the last thing he would wish to do would be to give notices which might prevent others from attempting, and probably succeeding, in their solution. There was, however, a problem because there could be some way of preventing disheartening and unnecessary duplication of work. Announcement of intention was hardly enough, and might indeed often be properly disregarded by the serious worker. Perhaps some kind of progress report from various laboratories, if it could be arranged, would help solve the problem.

PROFESSOR BERNAL suggested that any serious undertaking of structures by laboratories should be reported through *Acta Crystallographica*.

#### THE ABSORPTION OF X-RAYS

PROFESSOR M. VON LAUE (Max Planck Institute, Göttingen) gave a short address on "Recent investigations on the absorption of X-rays in crystals in the case of interference maxima." Although, in general, the refractive index has the same significance for X-rays as for visible light, the dynamical theory of X-ray diffraction shows that this is no longer so in the case of constructive interference,<sup>(15)</sup> and the same is true of the absorption coefficient. The data compiled in tables refer mainly to rays penetrating the crystal in directions other than those of constructive interference but, as deviations from the normal absorption process are not important except for very perfect single crystals, this restriction has almost escaped observation. Borrmann<sup>(16)</sup> has pointed out the defect in quartz.

If a divergent beam of X-rays from a copper target passes through a very perfect crystal of calcite of about

0.25 cm thickness and falls on a photographic film then, in general, the attenuation of the X-rays is so great that the crystal is practically opaque and there is no blackening of the film. However, there are positions in which the radiation fulfils the condition for constructive interference and intensity maxima corresponding to these positions are recorded on the film.

These effects may be explained in terms of the dynamical theory of X-ray diffraction<sup>(17)</sup> by substituting complex values for certain constants in the theory, as yet dealt with only as real quantities. A quantitative explanation of the phenomenon cannot be expected yet since the imaginary parts of these constants have not been measured or even estimated from the experimental results, but, nevertheless, the new theory seems to be readily applicable to recent observations of absorption phenomena and can provide a basis for further experiments.

#### CHEMICAL BONDS

In his evening lecture on "Chemical bonds—their shape and their behaviour," PROFESSOR C. A. COULSON (King's College, London) discussed modern views on bond lengths and the nature of chemical bonds. He indicated the importance of accurate measurements of electron densities by X-ray crystallographers in providing data for further advances in the theory.

#### GEIGER COUNTER TECHNIQUES IN X-RAY ANALYSIS

##### *Some Geometrical Factors affecting the Position and Shape of Powder Diffraction Maxima.*

The first paper in this session (Saturday morning, 3 December, 1949) was given by DR. A. J. C. WILSON (University College, Cardiff), who described work on the theory of the Geiger counter spectrometer which he had carried out in the Philips' Laboratories, Irvington-on-Hudson, New York. Ideally X-rays diverging from the source reunite, after diffraction by the specimen, to form a fine focus at the receiving slit, but in practice the conditions for such ideal focusing cannot be satisfied exactly and the diffraction maxima are displaced and unsymmetrically broadened. The effects of divergence of the rays in a plane perpendicular to the focusing circle have been discussed by Alexander<sup>(18)</sup> and may be largely eliminated by the use of Soller slits.<sup>(19)</sup> There are two main effects: that due to the finite height of the receiving slit and that due to the finite height of the irradiated part of the specimen. With a finite slit height  $h$  the maximum is smeared out over a range

$$\Delta\theta_1 = h^2 \cot 2\theta / 16R^2 \quad (5)$$

on the low angle side of  $\theta$  ( $R$  is the radius of the spectrometer). The displacement of the centre of gravity of the line is one-third of this amount. If the specimen is irradiated over a height  $s$  the diffraction maximum will be smeared out over a range of

$$\Delta\theta_2 = s^2(1 + \cos 2\theta)^2 \cot 2\theta / 16R^2 \quad (6)$$

also on the low angle side and, again, the displacement of the centre of gravity of the line is one-third of this. For ordinary dimensions ( $s \sim h \sim 1$  cm,  $R \sim 10$  cm) the coefficients of the trigonometrical terms are of the order of  $0.01^\circ$ , which would give very asymmetric lines and a total displacement of several tenths of a degree at low angles. By the use of Soller slits, however, the effective ratio of  $s$  (or  $h$ ) to  $R$  is reduced by a factor of five or ten and the effects of vertical divergence become small compared with those of horizontal divergence in conjunction with a flat specimen of finite absorption.<sup>(20, 21)</sup> It is ordinarily impracticable to shape the specimen to lie on the focusing circle as the curvature required is a function of  $\theta$ . A flat plate is therefore used and its divergence from the focusing circle produces a shift of the diffraction maxima to lower  $\theta$ . If the specimen is not effectively thin (i.e. either thin or of high absorption coefficient so that only the surface layers are effective) penetration of the X-rays into it also produces a displacement to lower  $\theta$ . For a consideration of the displacement of the maximum both these effects must be taken into account, but it is found that the effects on the centre of gravity of the maximum are simply additive and reflexion takes place over a range of  $\theta$  equal to

$$\Delta\theta_3 = (A^2/2R^2) \sin 2\theta \quad (7)$$

where  $2A$  is the length of specimen irradiated; the displacement of the centre of gravity is one-third of this amount. Absorption in the specimen (thickness  $t$ ) results in the diffraction maximum extending over a range of  $\theta$  equal to

$$\Delta\theta_4 = (t/R) \cos \theta \quad (8)$$

but the intensity of the low angle tail is usually much reduced by absorption. In this case the displacement of the centre of gravity is, approximately,

$$\Delta\theta_4 \simeq \sin 2\theta/4\mu R \quad (9)$$

where  $\mu$  is the absorption coefficient.

DR. WILSON also discussed the displacement of line peaks and the accuracy of lattice parameter measurements on a Geiger counter spectrometer and indicated that the errors due to displacement should vanish on extrapolation against  $\cos^2 \theta$  or  $\cos \theta \cot \theta$ . Since it is possible to find the zero of the angle scale of a Geiger counter spectrometer only by the use of a calibrating substance, the angles may all be in error by a constant amount due to the uncertainty in the angles of reflexion of the calibrating substance.<sup>(22)</sup> The fractional error in spacing introduced is proportional to  $\cot \theta$  so that any gross error in the zero of the angle scale would be detected by curvature of the extrapolation plot above  $80^\circ$ . A small error will produce an error in the extrapolated spacing of the order of  $10^{-4} \cot \theta \max.$  where  $\theta \max.$  is the Bragg angle of the highest measured reflexion.<sup>(23)</sup> There seems to be no reason why the Geiger counter spectrometer should not reach the same accuracy in the measurement of spacings as does the powder camera, but in order to surpass the latter some

method of locating the zero, or rather the  $90^\circ$ , point of the angle scale without using a calibrating substance must be devised.

### Applications of Geiger Counter Technique.

A paper by DR. P. T. CARTER and MR. I. R. HARRIS (Royal Technical College, Glasgow) discussed the application of Geiger counter technique to the examination of steel-making slags. Presenting the paper, DR. CARTER spoke of the importance of a knowledge of the oxidizing power and basicity of the refining slag and mentioned a number of physical methods which have been used in the endeavour to obtain a rapid and accurate measure of the basicity of a slag.<sup>(24), (25), (26), (27), (28)</sup> The basicity is usually expressed as the ratio  $\text{CaO}/(\text{SiO}_2 + \text{P}_2\text{O}_5)$  (the  $V$ -ratio) which may vary in normal practice from 1.5–3.0. The work described in the paper was undertaken in order to ascertain whether it would be possible to deduce the basicity of the slag from determination of the intensity of X-ray diffraction lines using a Geiger counter.

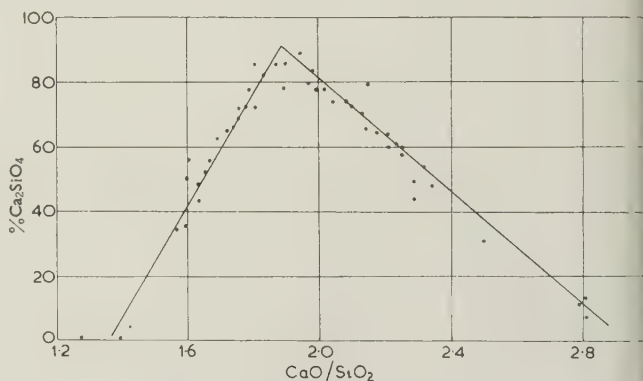


Fig. 1. Dicalcium silicate in slags

Of the components present in steel-making slags,<sup>(29), (30)</sup> CaO and  $\text{SiO}_2$  account for the greater part of the weight of the slag and these are largely combined as di- and tri-calcium silicates. For slags containing about 85%  $(\text{CaO} + \text{SiO}_2)$  the percentage of dicalcium silicate is a maximum at a  $\text{CaO}/\text{SiO}_2$  (weight) ratio of slightly less than 2 (Fig. 1). When slightly larger amounts of other constituents are present the amount of dicalcium silicate should be zero at high basicities, rise to a maximum at some point in the intermediate range, and fall again to zero at low basicities so that X-ray determination of the percentage of dicalcium silicate present appears to be a promising method of determining basicity.

Measurements were made with a Philips Geiger Counter Spectrometer using filtered copper radiation and taking counts over 15 sec intervals, the specimens being thin layers of powder mounted in depressions in Perspex slides. It has been shown that to reduce the mean deviation in the results to 10% a particle size of  $20 \mu$  or less is required,<sup>(31)</sup> and the quickest way of preparing samples was found to be crushing in a roll



usher and separation of the fines which pass through 300 mesh screen. With specimen thicknesses ranging from 0.02–0.12 cm absorption effects<sup>(32)</sup> were within the limits of experimental error. The intensity of a line occurring at  $2\theta = 41.1^\circ$ – $41.5^\circ$  (which could be due to either dicalcium silicate or tricalcium silicate or both) was chosen as a measure of basicity and Fig. 2 shows the results obtained. A comparison of Figs. 1 and 2 suggests that the line in question may be due to dicalcium silicate and indicates that there is sufficient promise in the Geiger counter method to warrant further investigation.

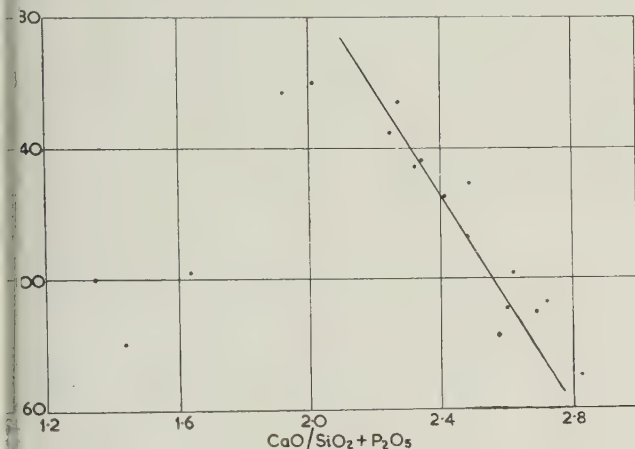


Fig. 2. X-ray measurement of basicity of slags

MR. D. A. OLIVER (B.S.A. Group Research Centre) asked if the alternative method of using a high-intensity X-ray beam and photographic film had been tried. Mr. Carter replied that with the Geiger counter technique a result could be obtained in 3 min, whereas the photographic method could not yet approach this speed and, in steel-making practice, time was very important. DR. J. W. JEFFREY (Birkbeck College, London) noted that great care was needed in the X-ray estimation of dicalcium silicate, it being possible in some cases for this substance to be missed even when present to as much as 30% of a mixture.

DR. R. F. HANSTOCK (High Duty Alloys Ltd.) gave a short account of the determination of pole figures for rolled aluminium, using a Geiger counter. He described the mounting of specimens ( $1 \times 0.085 \times 0.020$  in) and the Geiger counter and the need for accurate knowledge of absorption corrections,<sup>(33), (34)</sup> and also showed typical pole figures obtained with this apparatus.

#### Automatic Recording of X-ray Spectra.

MR. C. WAINWRIGHT (National Physical Laboratory) pointed out that the primary advantage of an automatic recording mechanism is the avoidance of the continuous close attention of an operator which is called for by the direct counting method and for some types of work, e.g. with mixtures involving a wide range of angles, it is the only reasonably practicable method. In providing for a given problem optimum conditions will be found,

influenced by the X-ray power available, the time constants of the electrical circuits, the extent to which fine detail must be shown, and an overall time schedule which may be determined by independent considerations. Although these conditions are not critical, too great a departure from them reduces the amount of information which is obtainable from the recorded spectrum.

DR. W. M. BARSS (Atomic Energy Research Establishment, Ministry of Supply) discussed the requirements of the electrical circuits used in the automatic recording of Geiger counter X-ray spectrometer data. Essentially two functions are required: (a) the generation of an electrical potential proportional to the rate of arrival of pulses from the Geiger counter, and (b) the continuous recording of this potential. In order to simplify the controls the experimental model described was arranged so that these two functions are isolated from each other and may be considered separately.

The rate-metering potential is provided by a simple integrating circuit consisting of a resistor  $R$  and a capacitor  $C$  in parallel into which are delivered equal charge increments  $q$  for each Geiger counter pulse. In order to be able to control the area under the voltage-time curve  $qR$  and the time constant of the circuit  $RC$  independently,  $R$  is kept constant,  $q$  and  $C$  being varied. It is the region of long time constant which is of interest, where the selection of  $RC$  is a compromise between desirable suppression of random fluctuations and undesirable distortion of apparent intensity curves. The average recorder deflexion is given by

$$\bar{V} = IRq \quad (10)$$

where  $I$  is the true average intensity. In general the instantaneous deflexion  $V$  is not equal to the average intensity value, the variance or mean square error being given by

$$s^2 = I/2RC \quad (11)$$

and the standard deviation, expressed as a fraction of the average deflexion, is

$$s/V = 1/(2\bar{V}RC)^{\frac{1}{2}} \quad (12)$$

However, when  $\bar{V}$  is determined by counting the number  $N$  of pulses in a period  $T$ , the fractional standard deviation is

$$s(N)/V = 1/(\bar{V}T)^{\frac{1}{2}} \quad (13)$$

Hence the accuracy at any point on the recorder curve is the same as would be obtained by counting the number of pulses received in time  $2RC$ .

When the period of integration vanishes the fractional standard deviation of  $\bar{V}$  obtained by integrating the curve over a given period  $T$ ,  $s(\bar{V}, T)$ , approaches  $s$ , whereas when the period becomes so long that the "memory" effect of the circuit is unimportant, it approaches  $S(N)$ .

$$\frac{s(\bar{V}, T)}{\bar{V}} = \frac{1}{(\bar{V}T)^{\frac{1}{2}}} \left[ 1 - \frac{RC}{T} \left( 1 - e^{-\frac{T}{RC}} \right) \right]^{\frac{1}{2}} \quad (14)$$

A graph of this quantity versus  $N$  for values of  $\bar{V}RC$  provides all the data necessary for finding the accuracy of intensity measurements to be expected under given experimental conditions, and hence provides a basis for the estimation of the smallest change in intensity which can be distinguished from purely random fluctuations in the curve traced for a constant intensity.

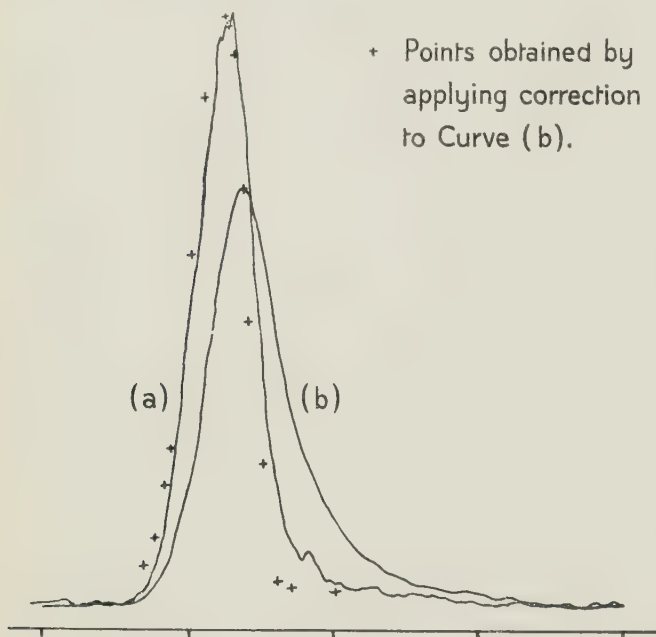


Fig. 3. Correction of a diffraction peak for integration-time error

(a) Scanning:  $\frac{5}{24}^\circ/\text{min}$  Chart:  $\frac{1}{2}$  in/min  $RC = 8.7 \text{ sec}$  (0.07 in).  
 (b) Scanning:  $\frac{5}{8}^\circ/\text{min}$  Chart: 2 in/min  $RC = 8.7 \text{ sec}$  (0.29 in).

The last part of the analysis concerns the unsymmetrical distortion of an ideal intensity curve which is produced by the time constant of the integrating circuit. The charge increments delivered with each pulse constitute a current which is divided between the resistance and capacity as follows

$$Iq = V/R + C\dot{V}$$

or

$$I = V + RC\dot{V} \quad (15)$$

when  $I$  and  $V$  are on the same scale ( $qR = 1$ ). The last term constitutes the correction, proportional to the rate of rise of the recorder trace, which must be added to the trace to give the true intensity. If  $RC$  is measured as a distance along the time axis of the chart,  $V$  may be obtained as the geometrical slope of the trace at the point for which the correction is desired. An actual diffraction peak corrected in this way is shown in Fig. 3. It is concluded that with a fast recorder tracing the output potential of the integrating circuit described it is possible to measure the intensity of diffracted X-rays with calculable accuracy and to make corrections for the distortion of diffraction peaks caused by the recording process.

### Line Widths and Intensities.

MR. W. H. HALL (University of Birmingham) described a spectrometer for the accurate measurement of Debye-Scherrer line widths and intensities using divergent monochromatic radiation and a geometric focusing method.<sup>(35)</sup> The intensity of the X-ray source is monitored continuously and errors due to variations in its output are automatically compensated by measuring the diffracted intensity not for a fixed time interval but for a fixed quantity of energy emitted by the tube. Measurements made on annealed and filed aluminium powders have shown that the measured integrated intensities agree with the accepted values to about 1%. Comparison of results obtained by graphical integration and by numerical integration showed that systematic differences occurred which could be attributed to diffuse tails extending for several degrees on both sides of the lines. Although these tails have only a small intensity ordinate, they extend so far that their integrated intensity may be several per cent of that of the line. They may be shown up on the trace as plotted by enlarging the intensity scale and compressing the angular scale, thus magnifying the background slope as shown in Fig. 4. The background intensity is so low that large statistical fluctuations are to be expected, but comparison of repeated measurements shows that the scatter of the points plotted is not completely random. In particular, small peaks which can be attributed to a second order component in the crystal reflected monochromatic beam occur, three low order ones being shown in Fig. 4.

The time involved in making these measurements depends on the accuracy desired. In the experiments described here each unresolved line was scanned in about an hour so that a complete spectrum requires about two days. Under these conditions, however, it is clear that very accurate results can be expected from Geiger counter apparatus.

In discussion DR. A. TAYLOR (Mond Nickel Co. Ltd.) observing that the  $K\alpha_3$  lines could not be seen in Mr. Hall's results, asked if Geiger counter techniques did give a better representation of line shapes than the usual photographic method. There was some discussion on this subject, but no definite reply to the question, although it was pointed out that the  $K\alpha_3$  lines might have been missed for a variety of reasons.

### Single Crystals.

DR. W. COCHRAN (Cavendish Laboratory, Cambridge) described the use of Geiger counters for single crystal measurements, confining his remarks to the accuracy obtainable in such measurements. He pointed out that the probable error of the quantity of interest to the crystallographer—bond length, electron density, etc.—is always proportional to the probable error of the measured structure factor so that every effort should be made to increase the accuracy of the experimental data. The Geiger counter, besides being almost 100% efficient for X-rays, is well adapted to the measurement of



Integrated reflexion intensity, and measurements have been made with copper radiation, monochromated when necessary by the use of balanced filters.<sup>(36)</sup> The method is less satisfactory when molybdenum radiation is used. Beyond about 100 counts/sec the relation between counting rate and X-ray intensity is not linear<sup>(37), (38)</sup> and, for a self-rectifying tube

$$N = \frac{N_0}{1 - N_0 K \tau} \quad (16)$$

where  $N$  = corrected,  $N_0$  = recorded counting rate,  $\tau$  = resolving time of apparatus,  $K$  = mean square intensity of source/(mean intensity of source)<sup>2</sup>.

In measuring integrated intensity the correction is more complicated since in addition to the short period variation

due to the pulsed nature of the source, there is a long period variation due to the varying reflecting power of the crystal. If the crystal moves with uniform angular velocity through a range  $\alpha$  (including the reflecting position) in a time  $T$

$$M = \frac{M_0}{1 - M_0 K_1 \tau / T} \quad (17)$$

where  $M$  = corrected,  $M_0$  = recorded counts in time  $T$  and  $K_1 = K \times$  mean square reflecting power over range  $\alpha$ /(mean reflecting power over range  $\alpha$ )<sup>2</sup>.

For measurements with molybdenum radiation the krypton filled counter is greatly superior to a photographic film. A reflexion sufficiently intense to enable 3 000 counts to be recorded in 1 min required an exposure time of about 1 hr to give a density ten times that just visible, only 5% of the radiation being absorbed in the emulsion.

In addition to the time factor, the Geiger counter method has been shown to be more accurate than the method of estimating intensities by eye. Two independent measures of structure factors by Geiger counter methods agree to about 3% and part of this variation is probably due to the approximate nature of the absorption correction.<sup>(39)</sup> These structure factors agree to about 10% with those estimated by eye. A final test of the accuracy of the counter measurements comes from the agreement between observed and calculated structure factors. In a particular case these values agreed to 6%, which compares with the figures of 20% obtained when the structure of the compound (adenine hydrochloride) was worked out using photographic data.<sup>(40)</sup> The use of the Geiger counter opens up a new field of investigation in crystal structure analysis as it should make possible the detection of electron densities of the order of  $0.1 e/A^2$  or  $e/A^3$ .

MR. P. B. HIRSCH (Cavendish Laboratory, Cambridge) spoke of the use of Geiger counter technique in a micro-beam study of the mosaic structure of crystals, the position of the counter being fixed and the crystal traversed parallel to its own surface.

By measuring the intensity of X-rays reflected from planes inclined to the surface of a crystal, the thickness of surface layers on crystals may be determined, values of the intensity measured by the Geiger counter technique being considerably more accurate than those measured photographically. The crystal is irradiated with a monochromatic beam of appreciable divergence so that the

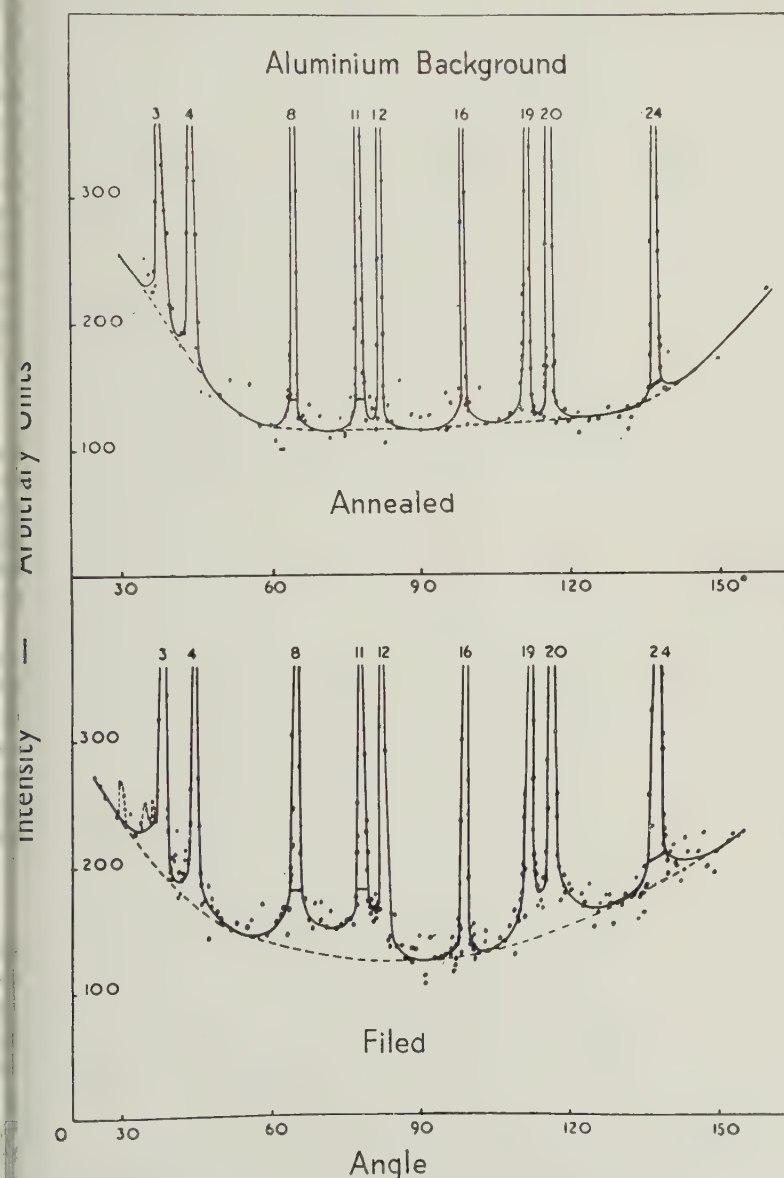


Fig. 4. Recorded X-ray spectra from aluminium

intensity of reflexion from the stationary crystal is proportional to the integrated reflexion.

Another use of Geiger counters is to align the lead glass capillaries used as collimators in the microbeam technique. This method could also be used with ordinary collimators, but DR. COCHRAN suggested that a photocell could be used for this purpose.

#### Geiger Counter Design.

Although the available X-ray intensity is, in general, limited yet for reasons of statistical accuracy as high a counting rate as possible is required so that it is important to consider the design of the actual counter tube. DR. U. W. ARNDT (University of Birmingham) pointed out that the principal demands made on a counter are: (1) high quantum efficiency; (2) high stability over prolonged periods; (3) either accurate linearity of response or the possibility of making corrections for counting losses at high intensity.

Because of the presence of a region immediately behind the counter window where no counts are produced, the filling pressure of the counter cannot be increased indefinitely since a point is reached when the increased absorption in the lead space more than makes up for that in the active volume. Also, in order to keep the recovery time of the counter (which is proportional to the filling pressure) as low as possible, too high a pressure is undesirable. For an argon filled counter of typical dimensions no advantage is gained by filling to a pressure of more than about 60 cm if the counter is to be used with  $\text{Cu } K\alpha$  radiation or more than 35 cm for  $\text{Fe } K\alpha$ .

To ensure stability the counter must be baked at an elevated temperature before filling and soft waxes and alkali or alkaline earth metals avoided in the construction. The probability of a spurious count is greatest just before the arrival of the positive ion sheath at the counter walls<sup>(41)</sup> and such pulses may therefore be suppressed by the use of a quenching circuit.<sup>(42), (43)</sup> Since it is mainly the spurious pulses which are temperature dependent, the use of such a circuit largely eliminates effects of temperature variations.

The recovery time of a counter is given by

$$t_r = \frac{Pr_c^2 \log_e(r_c/r_w)}{2Vk} \quad (18)$$

where  $P$  = filling pressure;

$r_c$  = cathode radius;

$r_w$  = wire radius;

$V$  = operating voltage;

$k$  = ion mobility.

Since the only readily available parameter in this formula is the cathode diameter it is advisable to make the counter diameter as small as possible. Characteristics of some typical counters are given in the accompanying table.

#### Characteristics of three counters

	A	B	C
Diameter	1 in	$\frac{3}{4}$ in	$\frac{1}{2}$ in
Pressure, cm mercury	12	70	30
Operational voltage	1 000	1 430	1 300
Plateau length	250	150	150
Plateau slope	0.06	0.08	0.04
Relative sensitivity	1.0	5.5	3.7
Recovery time, $\mu$ sec	375	800	180
Dead time	120	170	100

A. Counter by All Tools Ltd. B. Experimental Counter by The General Electric Co. Ltd. C. Counter by 20th Century Electronics.

Counting losses can be computed with far greater precision when a quenching circuit is used since the accurately known quench time now takes the place of the ill-defined recovery time of the counter. The actual insensitive time must be multiplied by a form factor which arises owing to the fact that the counting rate at the peak of the half-cycle is far greater than the average counting rate.<sup>(44), (45)</sup>

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## ORIGINAL CONTRIBUTIONS

### Mechanical Models for the Representation of Visco-Elastic Properties

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Models of varying complexity, built of spring and dashpot elements, are frequently used as a simple means of expressing the relation between a particular component of stress and the corresponding component of strain in a visco-elastic material. It is shown that any such model is equivalent to two alternative spring-dashpot models of canonic form. The exclusive use of models in either of these canonic forms is desirable, since it facilitates the interpretation of behaviour and leads to economy in the number of spring and dashpot elements employed.

#### INTRODUCTION

Mechanical models are now very generally used for the representation of visco-elastic properties. In such models, which are built of spring and dashpot elements, the extension and applied force are taken to represent respectively a component of strain and the corresponding component of stress in the material. While this method

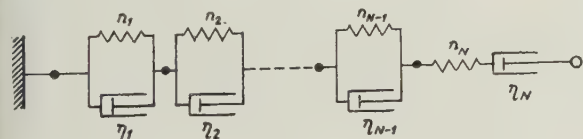


Fig. 1. Typical "canonic" form of model

of representation is precisely equivalent to writing down a differential equation relating stress, strain and time, it has a considerable practical advantage over the latter method in that the behaviour of a material in a given deformation process can often be inferred from a glance at the appropriate model, but not so easily from the differential equation.

The models most frequently used are of the types

shown in Figs. 1 and 2; a satisfactory representation being often obtained with the use of a small number of elements. For every model belonging to one of these types, a model of the other type can be constructed

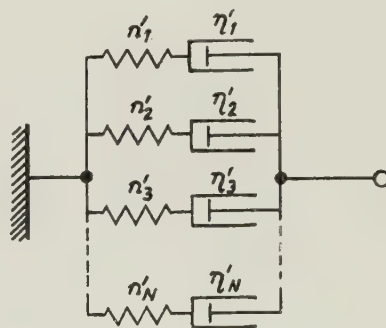


Fig. 2. Another typical "canonic" form of model

having the same number of spring-dashpot pairs  $N$ , which shows identical mechanical behaviour. Bilmes<sup>(1)</sup> has pointed out this equivalence for the case in which  $N = 2$ , and has given the formulae relating the constants  $n_1, n_2, \eta_1, \eta_2$  in one model with the constants  $n'_1, n'_2,$

$\eta'_1, \eta'_2$  in the equivalent model of the other type; while Alfrey<sup>(2)</sup> has shown that the equivalence holds for all values of  $N$ , and has given methods for computing one set of constants from the other.

When a complex visco-elastic material is under investigation, and it becomes apparent that a large number of elements will be necessary in the representative model, the following questions arise: (1) Can the behaviour be represented by models of these two simple types, or must a more complex model be used, such as the example shown in Fig. 3? It is shown below that any model is equivalent to models of the two "canonic" forms shown in Figs. 1 and 2 (containing suitable numbers of springs and dashpots), so that it is unnecessary to construct complex models. (2) Is economy in the number of springs and dashpots obtained by using the canonic forms rather than a complex model? It is shown below that a great saving in the number of elements often results from a use of the canonic forms, and rules are given for deducing the number of

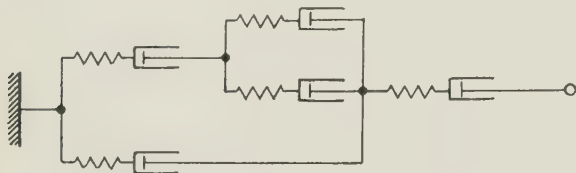


Fig. 3. More complex model

elements in the canonic forms which are equivalent to a given model.

It must be remembered that when a model has been found which gives a satisfactory representation of the observed behaviour of a material, the representation must be regarded as approximate rather than exact since more accurate or more extended observations will almost certainly show divergences in behaviour. Such divergences will require the addition of further elements to the model, and an advantage of constructing the latter in either of the types shown in Figs. 1 and 2 is that such additions can be made without altering the form of the model. The case of rubbers, for instance, has frequently been considered using a model of the second canonic form, and it is found that a large number of spring-dashpot pairs is necessary for accurate representation (Treloar<sup>(3)</sup>). This has led to the idea that such materials are correctly represented by a continuum whose properties are the limiting properties of a model as the number of elements is made very large, the constants varying continuously from pair to pair: a generalization of the model representation which can only be made on the basis of a canonic form.

#### THE ELECTRICAL ANALOGY

The general proof that any model is equivalent to a model of either canonic form does not appear in the literature, although Kuhn<sup>(4)</sup> has considered the equi-

valence between any model and one of the form shown in Fig. 2 as regards relaxation at constant deformation only. The general proof is long and need not be given as an analogous proposition in electrical network theory has been proved elsewhere. In order to make use of this result of electrical theory, it is necessary to establish an exact analogy between a two-terminal network containing resistors and capacitors and a mechanical model containing springs and dashpots. Now the behaviour of a model is conditioned by relations for its spring elements of the type

$$\text{Force} = n \times \text{Extension}$$

and relations for its dashpot elements of the type

$$\text{Force} = \eta \times \text{Rate of extension}$$

together with the conditions that the algebraic sum of the forces at each vertex shall vanish, and that the sum of the extensions of the elements traversed by every path from the fixed wall to the movable terminal shall be the same. On the other hand, the behaviour of a two-terminal resistor-capacitor network is conditioned by relations for its resistance elements of the type

$$\text{Current} = 1/R \times \text{Potential difference}$$

and relations for its capacitance elements of the type

$$\text{Current} = C \times \text{Rate of change of potential difference}$$

together with the conditions that the algebraic sum of the currents at each vertex shall vanish, and that the sum of the potential differences across the elements traversed by every path joining the two terminals shall be the same. Thus for any model, an analogous electric network may be constructed by replacing the fixed wall by a terminal, each spring by a resistor  $R = 1/n$ , and each dashpot by a capacitor  $C = \eta$ ; and the relation between force and extension in the model is then identical with that between current and potential difference in the electric network.\*

Now it has been shown by Cauer<sup>(7)</sup> that any two-terminal resistor-capacitor network is equivalent to two alternative networks of canonic form which are the electrical analogues of the models shown in Figs. 1 and 2 (obtained by replacing springs by resistors and dashpots by capacitors). Actually, in Cauer's work (of which an account has also been given by Guillemin<sup>(8)</sup>) the equivalence is only shown to exist for alternating currents in the steady state, but since the same equivalence exists

\* Alfrey and Doty<sup>(5)</sup> mention a more obvious analogy, namely that between force and potential difference, rate of extension and current, springs and capacitors, dashpots and resistors. While this is correct for individual elements, it breaks down for networks, since the vertex conditions involve force and current analogously and the continuity conditions involve extension (or rate of extension) and potential difference analogously. The difficulty may be got over, as indicated by Whitehead,<sup>(6)</sup> by replacing springs by capacitors and dashpots by resistors and constructing the dual of the circuit so obtained. The relation between force and rate of extension is then the same as that between potential difference and current in the dual; but such a treatment is much more cumbersome than that given in the text above.



Whatever the frequency of these currents, it must exist in all circumstances. The integer  $N$  which determines the complexity of these canonic networks is given by the relation

$$N = B - V + 1$$

where  $B$  is the number of independent branches in the original network and  $V$  the number of its vertices.

#### CANONIC FORMS FOR MECHANICAL MODELS

By transferring Cauer's result to spring-dashpot models, it can be seen that any such model has equivalent canonic forms of the types shown in Figs. 1 and 2, and the integer  $N$  can be found by inspection using the rule given above for electric networks. Thus, for the example shown in Fig. 3,  $B = 5$  and  $V = 3$  so that  $N = 3$ , and the canonic forms only contain six elements opposed to ten in the original model. It must be noted in reckoning the value of  $B$ , that if the spring and dashpot on the right-hand side of Fig. 3 were absent, the rod joining the right-hand vertex to the terminal would still rank as a branch. On the other hand, the section between the wall and the left-hand vertex never ranks as an independent branch, even if it contains a spring and dashpot (since these elements could be transferred to the right-hand branch without affecting the behaviour of the model).

In certain cases, one or even two elements may be missing from the canonic forms, so that the total number of elements in the latter is less than  $2N$ . This is not the case, however, for a model such as that shown in Fig. 3 in which every path from the wall to the terminal traverses at least one spring and one dashpot (showing that the material represented is a liquid which exhibits an instantaneous elastic recovery on removal of stress). For in the canonic forms also, every path must traverse at least one spring and one dashpot; and an inspection of Figs. 1 and 2 shows that this can only be the case if all springs and dashpots are present. The state of affairs is different, however, if it is possible to find a path from wall to terminal which only traverses dashpot elements (the model now representing a liquid which shows no instantaneous elastic recovery). For in the corresponding model of the first canonic form the right-hand spring must be absent, and one of the springs in the model of the second canonic form must be absent, so that number of elements is  $2N - 1$ . On the other hand, it may be possible to find a path which only traverses spring elements (the model now representing a solid), so that the right-hand dashpot in Fig. 1 and one of the dashpots in Fig. 2 must be absent, and the number of elements is again  $2N - 1$ . The remaining possibility is that of a model in which one path can be found which only traverses springs and another which only traverses dashpots (the model representing a solid which would require an infinite stress to produce an instantaneous deformation). In this case, both spring and dashpot are absent from the right-hand side of Fig. 1, and one

spring and one dashpot are absent from different branches of Fig. 2, the number of elements being  $2N - 2$ .

In conclusion, it is necessary to emphasize that these rules for the reduction of a model to its canonic forms

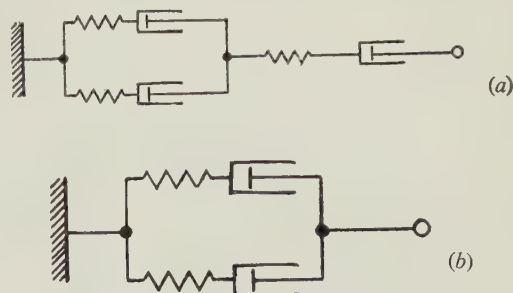


Fig. 4. Simplification of model

are of importance even in very simple cases. For example, they enable the model shown in Fig. 4(a) to be reduced at once to the much simpler model of Fig. 4(b), a rather surprising result. Again, by reducing the model shown in Fig. 5(a) to the canonic form of the type shown in Fig. 2, and noting that one spring must be absent in the latter, the equivalent model of Fig. 5(b) is obtained. (Actually Fig. 5(a) is already in the canonic

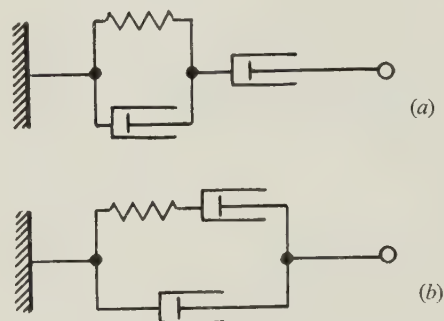


Fig. 5. Reduction of model to "canonic" form

form of Fig. 1.) This case is mentioned here, because it is sometimes erroneously stated that the models of Fig. 5 exhibit different mechanical behaviour.<sup>(9)</sup>

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# The Formation and Crystal Structure of Silicon Carbide

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The presence of cubic  $\beta$ -SiC has been identified by X-ray photographs when graphite and silicon are heated together at temperatures as low as 1 150° C, and when vitreous silica is heated with graphite the carbide is formed at 1 450–1 475° C, probably by a vapour phase reaction. No matter how the starting materials may be varied in nature and in proportion, face-centred cubic carborundum is always formed, unless the temperature is in the region of the decomposition point. Near 2 000° C face-centred cubic carborundum begins to decompose, as shown by the presence of graphite lines in an X-ray photograph, and lines corresponding to the hexagonal modification II appear. Light green commercial carborundum shows faint graphite lines when heated at 2 000° C, while at 2 050° C it turns black and a large amount of macrocrystalline graphite is formed. The long soaking periods, high vapour pressure, and high temperature conditions occurring in commercial processes favour the formation of large crystals of modification II.

The evaluation of the Debye-Scherrer photographs from commercial silicon carbides shows that the elementary tetrahedra which form the basis of the structures are slightly distorted, being expanded in the  $c$  direction and compressed in the direction of the  $a$ -axis. A possible wurtzite type, not so far reported in the literature, was looked for, but without success.

While the X-ray literature indicates that there is a number of crystallographic modifications of silicon carbide<sup>(1-7)</sup> a critical analytical investigation by K. Arnt and E. Hausmann<sup>(8)</sup> excludes the possibility of the existence of any formula for silicon carbide other than SiC. They also reject the existence of various oxy-carbides reported by earlier workers. As early as 1911, H. W. Gillett<sup>(9)</sup> pointed out that the values given by different workers for the formation and decomposition temperatures of crystalline carborundum varied greatly. Formation temperatures for SiC ranging from 1 220° C to above 2 000° C are quoted. Most authors agree on a decomposition temperature of approximately 2 200° C, although 2 700° C has been reported.<sup>(10)</sup> Density measurements also reveal large variations.

H. Ott<sup>(1)</sup> has shown that SiC exists in five crystallographic modifications, one of which is the so-called "amorphous carborundum." Four of the modifications are illustrated in Fig. 1. They are built up of tetrahedral units of four carbon atoms which surround each silicon atom in such a way that each carbon atom is also surrounded tetrahedrally by four silicon atoms. "Amorphous carborundum" modification IV in Ott's notation and  $\beta$ -SiC according to Thibault,<sup>(3)</sup> is really a misnomer for the cubic form of carborundum. It obtains its name from the fact that it is usually found in the form of a very fine powder, and it is not amorphous in the sense that it possesses a random structure like a glass.<sup>(11)</sup> From goniometric measurements on single crystals, Thibault<sup>(3)</sup> discovered a further two modifications, the tetrahedral atomic arrangements of which were shown by Ramsdell<sup>(4, 5, 6)</sup> to be closely

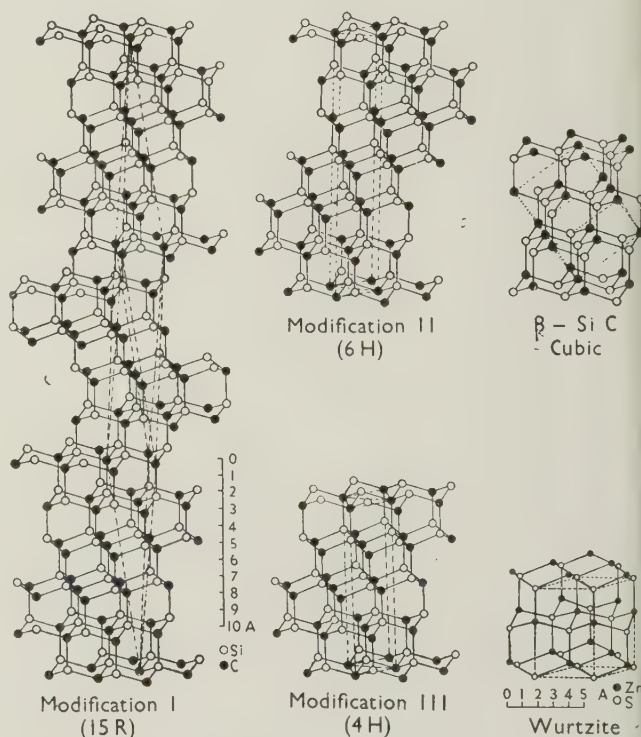


Fig. 1. The crystal structures of SiC (after H. Ott)

related to those in the structures originally determined by Ott.

The cubic form has essentially the same type of structure as zinc blende ZnS, or diamond, but with silicon atoms occupying the centres of tetrahedra which are normally occupied by carbon atoms. We may look upon the cubic structure as made up of

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† Now at Goodlass Wall and Lead Industries, Ltd.



parallel sheets of tetrahedra which lie perpendicular to the trigonal axis of the cube. The individual tetrahedra are also set in a direction parallel to each other. The various hexagonal modifications can be derived from the cubic variety by rotating the tetrahedra in alternate sheets so that they lie in parallel or anti-parallel directions. This form of multiple twinning about the hexagonal axis is in some ways analogous to the formation of a "mistakes" lattice in hexagonal cobalt,<sup>(12, 13)</sup> and may, in certain cases, give rise to similar diffraction effects. The unit cell dimensions and space groups of the various forms which have so far been isolated are given in Table 1.

Tone<sup>(14)</sup> has examined a large number of silicon carbide varieties by means of Laue photographs. He has shown that intermediate patterns exist in the photographs and states that it seems possible to trace a gradual transition from one pattern to another by the appearance or disappearance of various diffraction spots. No quantitative evaluation of the photographs was indicated. We have attempted to prepare laboratory specimens of the various forms of carborundum which have been found to exist in the commercial samples of the above investigators and to identify them by means of X-ray analysis. Commercial preparations kindly supplied by the Carborundum Co. Ltd., and the Universal Grinding Wheel Co., of Stafford, were also examined, and these gave most interesting results. The influence of temperature and of changes in the materials used in the preparation of the laboratory samples were carefully investigated and accurate formation and decomposition temperatures were obtained. A most interesting feature

in the formation of the carbide under laboratory conditions was that the cubic form,  $\beta$ -SiC, was always obtained, no matter how the starting materials were varied. Only when the cubic variety was heated almost to the decomposition temperature were hexagonal modifications produced.

#### PREPARATION AND X-RAY EXAMINATION OF THE SPECIMENS

The first three samples were prepared in air, in a graphite crucible in an induction furnace. All subsequent preparations were made in evacuated carbon tube furnaces. The samples were usually in the form of a powder and were placed in a pure graphite test tube which was lowered into the carbon heater element. New test tubes were used for critical specimens. Temperature measurements were made with an optical pyrometer of the disappearing filament type, and also by means of a Féry radiation pyrometer. The carbon tube furnaces proved much steadier in operation than the induction furnace and were very easy to control. Consequently, accurate temperature measurements were obtainable.

All the preparations were finely ground to sieve through fine copper gauze with 350 meshes to the inch, and then made into cylindrical rods, 0.3 mm in diameter, with dilute glue solution as a binder. Debye-Scherrer powder photographs were taken from the rods in 14 cm and 19 cm diameter cameras with Co K $\alpha$  radiation, using a Metropolitan-Vickers Raymax tube and an experimental rotating anode X-ray tube con-

Table 1. *The relationship between the forms of SiC*

SiC designation <i>Ott, Thibault-Ramsdell</i>		Space group	Lattice parameters <i>Absolute Angström units</i>	Axial ratio <i>c/a</i>	Molecules per hexagonal cell	Authority for parameter data
Undiscovered Type	Wurtzite $\alpha$ -SiC	$C_{6v}^4$ -C6mc	$a = 3.0817$ (Predicted) $c = 5.0394$ values	$1.0 \times 1.63529$	2	Taylor and Laidler
Mod. IV	$\beta$ -SiC	$T_d^2$ -F $\bar{4}3m$	$a_w = 4.3590$	$1.5 \times 1.633$	4	Taylor and Laidler
Mod. III	4H $\alpha$ -SiC III	$C_{6v}^4$ -C6mc	$a = 3.079$ $c = 10.254$	$2.0 \times 1.633$	4	Thibault
Mod. II	6H $\alpha$ -SiC II	$C_{6v}^4$ -C6mc	$a = 3.0817$ $c = 15.1183$	$3.0 \times 1.63529$	6	Taylor and Laidler
Mod. I	15R $\alpha$ -SiC I	$C_{3v}^5$ -R3m	$a = 3.079$ $c = 37.78$ $a_{rh} = 12.72 \alpha = 13^\circ 54.5'$	$7.5 \times 1.633$	15	Thibault
—	21R $\alpha$ -SiC IV	$C_{3v}^5$ -R3m	$a = 3.079$ $c = 52.88$ $a_{rh} = 17.72 \alpha = 9^\circ 58'$	$10.5 \times 1.633$	21	Thibault
—	33R $\alpha$ -SiC VI	$C_{3v}^5$ -R3m	$a = 3.079$ $c = 83.10$ $a_{rh} = 27.75 \alpha = 6^\circ 21.5'$	$16.5 \times 1.633$	33	Thibault
Mod. V	51R $\alpha$ -SiC V	—	$a = 3.079$ $c = 128.434$ $a_{rh} = 42.85 \alpha = 4^\circ 07'$	$25.5 \times 1.633$	51	Ramsdell
—	87R $\alpha$ -SiC	$C_{3v}^5$ -R3m	$a = 3.079$ $c = 219.094$ $a_{rh} = 73.011 \alpha = 2^\circ 25'$	$43.5 \times 1.633$	87	Ramsdell

<i>Materials used</i>	<i>Conditions</i>	<i>Experimental observations</i>	<i>X-ray data</i>
2 g Ceylon graphite 2 g crushed quartz	2 h at 1600° C in induction furnace	Feathery mass of vitreous SiO <sub>2</sub> on top of main charge. Main charge small light green crystals	Face-centred cubic SiC + a little unchanged graphite. Feathery mass of SiO <sub>2</sub> gave the broad band of vitreous silica.
1 g Ceylon graphite 5 g crushed quartz	3 h at 1500° C in induction furnace.	Feathery SiO <sub>2</sub> on top of charge. Main charge light green hard crystals. Globule of clear SiO <sub>2</sub> adhering to bottom of crucible.	Face-centred cubic SiC for main charge.
3 g Ceylon graphite 1 g crushed quartz	3 h at 1800° C in induction furnace.	Charge black, but contained hard particles	Face-centred cubic SiC + unchanged graphite
0.8 g Ceylon graphite 0.4 g silicon (crystalline)	2 h at 1800° C	Film formed over charge at about 1500°. Some light green crystals observed in main charge by microscope.	Face-centred cubic SiC + unchanged graphite.
0.25 g Ceylon graphite 1 g silicon	1 h at 1300° C. 1 h at 1300–1400° C. 2 h at 1800° C.	Charge had much greener appearance than previous charge. Sample wet oxidized to remove unchanged graphite.	Face-centred cubic SiC + a trace of unchanged graphite.
0.5 g Ceylon graphite 2 g silicon	3 h at 1500° C.	Charge green in appearance. Sample wet oxidized to remove excess graphite.	Face-centred cubic SiC (after wet oxidation).
1 g Ceylon graphite 2 g silicon	3 h at 1300° C.	Charge dark in appearance.	Unchanged graphite + unchanged silicon. Face-centred cubic SiC present, but spectrum lines not as sharp as from set of higher temps.
1 g Ceylon graphite 2 g silicon	3 h at 1200° C.	Charge black but contained some hard particles.	As above, but with the face-centred cubic SiC lines not so well defined.
1 g Ceylon graphite 2 g silicon	3 h at 1150° C.	Charge as above.	Mainly unchanged Si and C, but with a little face-centred cubic SiC formed. High order lines not visible. Low order lines broad, i.e. the SiC is approaching colloidal dimensions.
1 g active charcoal 3 g silicon	3 h at 1800° C.	Charge appeared greyish-green.	Face-centred cubic SiC + small amount of unchanged carbon. The latter shows great increase in particle size.
0.6 g charcoal 2 g silicon	3 h at 1200° C.	Charge grey in colour.	Face-centred cubic SiC quite sharply defined (Crystallite size greater than 1 000 Å). Unchanged silicon also present.

structed by one of us at The English Electric Co. Ltd., Stafford.<sup>(15)</sup> In certain cases when high resolution of a selected group of lines was required, photographs were taken with Fe K $\alpha$  radiation. The angle of the 19 cm diameter camera from which the lattice parameters were deduced was calibrated using clear quartz as a standard.<sup>(16, 24)</sup> Where necessary, the samples were chemically analysed to establish the formula of the carbide.

The results of various methods of preparing silicon carbide and of heating the laboratory and commercial varieties in vacuum to high temperatures are summarized in Table 2.

It was found that when compressed pellets of silicon-graphite mixtures were heated, the reaction was so vigorous that the charge was blown out of the furnace, and it was necessary to use uncompacted mixtures of powder in order to control the reaction rate. Experiments were also carried out in the temperature range 1 600° to 1 800° C, with  $\frac{1}{4}$  in diameter pure graphite rods embedded in a compact mass of fine silica powder

in the hope that a layer of silicon carbide would form on the surface of the graphite. In most cases, it was found that the silica had fused together into a solid mass and the graphite rod had been reduced to about half its diameter with no silicon carbide either on the surface of the rod or on the surface of the silica. Instead, it was found that silicon carbide crystals had deposited some distance away on the cooler zones of the graphite container. Only occasionally would crystals of silicon carbide be found on the surface of the graphite rod. When pure silica was used, these crystals would be pale green in colour, but quite black if ordinary sand contaminated with iron oxide was employed. The above experiments indicate that silicon and graphite react together in the solid state at as low a temperature as 1 150° C. It would appear, however, that the carbide, when prepared from a mixture of silica and graphite, is formed via a vapour phase reaction at about 1 450° C.

The powder diagrams show quite conclusively that below 2 000° C only the cubic form,  $\beta$ -SiC, is formed. Above this temperature and in the region of 2 200° C



## formation of carborundum

Materials used	Conditions	Experimental observations	X-ray data
10 g Ceylon graphite 10 g sand	2 h at 1800° C.	Film of amorphous SiO <sub>2</sub> formed over charge and also a hard "collar" of face-centred cubic SiC between the film and main charge.	Main charge face-centred cubic SiC + a little unchanged graphite.
10 g coke 10 g sand	3 h at 1800° C.	Film formed over charge at approximately 1700° C.	Face-centred cubic SiC + a little carbon.
Commercial charge 10 g coke 5 g sand 0.50 g sawdust 0.15 g NaCl	Carbonized at 900° C for 1 h, then 4½ h at 1900° C.	Carbonization first in air at 900° C to get uniform charge for furnace.	Face-centred cubic SiC + a little carbon.
Amorphous vitreous SiO <sub>2</sub> 10 g Ceylon graphite 10 g ball compressed pellet	3 h at 1250–1290° C.	Charge grey in colour.	Cristobalite + a little unchanged graphite. No SiC formed.
Amorphous vitreous SiO <sub>2</sub> 10 g Ceylon graphite 10 g ball compressed pellet	2 h at 1450° C.	Charge grey in colour.	Trace of SiC (?), cristobalite, and trace of carbon.
Amorphous vitreous SiO <sub>2</sub> 10 g Ceylon graphite 10 g ball compressed pellet	3 h at 1475° C.	Charge light grey in colour.	Face-centred cubic SiC present with cristobalite and trace of carbon.
Face-centred cubic carborundum prepared from 0.25 g Ceylon graphite 1 g Si heated to 1800° C for 2 h	3 h at 2100° C.	Sample assumed much darker colour but still abrasive.	Face-centred cubic SiC with strong graphite pattern. Lines from hexagonal SiC appearing.
10 g Ceylon graphite 2.5 g Si	1 h at 1800° C, 1 h at 2000° C, 2 h at 2300° C.	Film of amorphous material formed near top of tube. "Collar" of face-centred cubic SiC formed above main charge in cooler part of tube.	Main charge hexagonal SiC + face-centred cubic SiC + graphite.
Face-centred cubic SiC from "commercial" charge at 1900° C for 4½ h	2 h soaking up to 2400° C.	Product contained much free graphite.	Hexagonal SiC lines still present. Large amount of graphite.
Face-centred cubic SiC from "commercial" charge at 1900° C for 4½ h	4 h at 2200° C.	As above.	Hexagonal SiC lines and much stronger graphite lines.
Commercial hexagonal SiC from the Carborundum Company Type 6H	2 h at 2050° C.	No change in appearance.	No change in the hexagonal SiC pattern, but faint carbon line appearing.
Light green commercial hexagonal SiC. Type 6H	2 h at 2050° C.	Charge went black.	Hexagonal SiC and strong graphite pattern.

The cubic form partially transforms into the hexagonal variety, modification II of Ott (6H in Ramsdell's notation), after heating for four hours. Evidently there is also some decomposition at this higher temperature for graphite lines also make their appearance in the powder photographs. These graphite lines are quite sharp, indicating a growth of the carbon decomposition product into crystals larger than  $10^{-5}$  cm in size. In the process, the more volatile silicon evaporates off leaving the carbon behind. However, the carborundum residue still has the stoichiometric composition SiC even though the crystal symmetry is changed.

The complete conversion of the cubic form into a hexagonal polymorph necessitates a much longer heating period than four hours. In large-scale commercial processes the period of heating is of the order of 36 hours and temperatures of 2200° C are reached. It is probably for this reason and on account of the higher vapour pressures which are attained that the commercial forms of carborundum are almost invariably hexagonal. The majority of the commercial samples we have been able

to examine show the same structure, modification II, whatever their colour or place of origin, and only a comparative few gave more complex patterns indicative of larger unit cells.

The purest commercial samples and the extremely pure laboratory products are a very pale green in colour. Chemical analysis shows them to be at least 99.8% SiC. Variations in colour ranging from the palest green to jet black with intermediate yellows, browns, greys, and purples are to be found in the commercial products. These colours are probably caused by traces of impurity, mainly iron, in the lattice, and although the lattice parameters and the intensities of the diffraction spectra seem to be in no way affected by the coloration, great variations can be obtained in the electrical characteristics. In the course of preparation, the addition of a little common salt ensures removal of the iron as a volatile chloride. Some of the lower grade commercial varieties give extra Debye-Scherrer reflexions due to the presence of admixed crystalline impurities. A selection of powder photographs from various forms of

silicon carbide taken with Co  $K\alpha$  radiation is given in Fig. 2, and illustrates the increasing complexities which occur as the unit cell is enlarged.

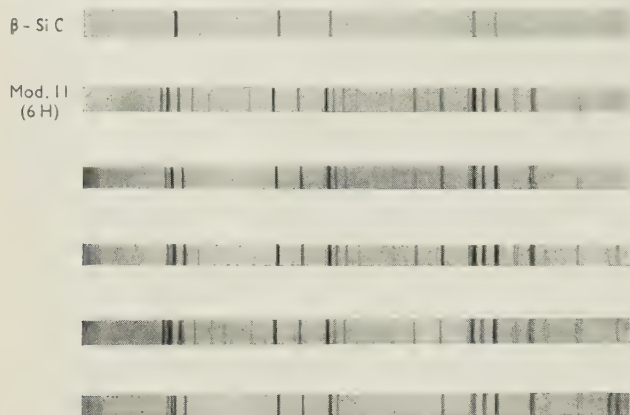


Fig. 2. Powder patterns of different forms of SiC. Co  $K\alpha$  radiation, 19 cm camera. Rotating anode tube. (15) Exposure 1 hr at 80 mA and 40 kV peak

#### THE LATTICE PARAMETERS AND STRUCTURE OF SiC MODIFICATION II

A careful inspection of the powder photographs of commercial carborundum showed that some of the lines were slightly broadened and others were quite definitely doubled. This occurred for lines which fell

in positions corresponding to lines from the cubic variety. The most noticeable doubling occurred with the groups  $10\bar{1}14$ ,  $20\bar{2}10$ ,  $21\bar{3}2$ ; and  $11\bar{2}12$ ,  $21\bar{3}4$  which correspond in position to reflexions 330 and 420 from the cubic form. It seemed at first as if the commercial carborundum was, in reality, a mixture of hexagonal modification II and cubic  $\beta$ -SiC. Careful examination of several photographs from different samples showed exactly the same phenomenon without revealing any changes in the relative intensities or positions of the spectrum lines. This appeared to rule out the possibility of a mixture and attention was therefore directed to checking the structure and the lattice parameters.

In this connexion it should be mentioned that Thibault<sup>(3)</sup> specifically states that lines due to common planes were always *strictly coincident* on the powder photographs. This lack of resolution was clearly caused by the use of a small camera of only 57.3 mm diameter and Cu  $K\alpha$  radiation.

The  $\sin^2 \theta$  values of the diffraction lines from a commercial light green carborundum were obtained, using Co  $K\alpha$  radiation and the 19 cm diameter Debye-Scherrer camera, and the values so obtained were compared with those computed from the precision determination of the parameters of modification II obtained by G. Borrmann and H. Seyfarth.<sup>(17)</sup> These workers used Cu  $K\alpha$  and Zn  $K\alpha$  radiations and employed the mixture method with diamond powder as the reference standard. They did not report the doubling of the spectrum lines. Their lattice para-

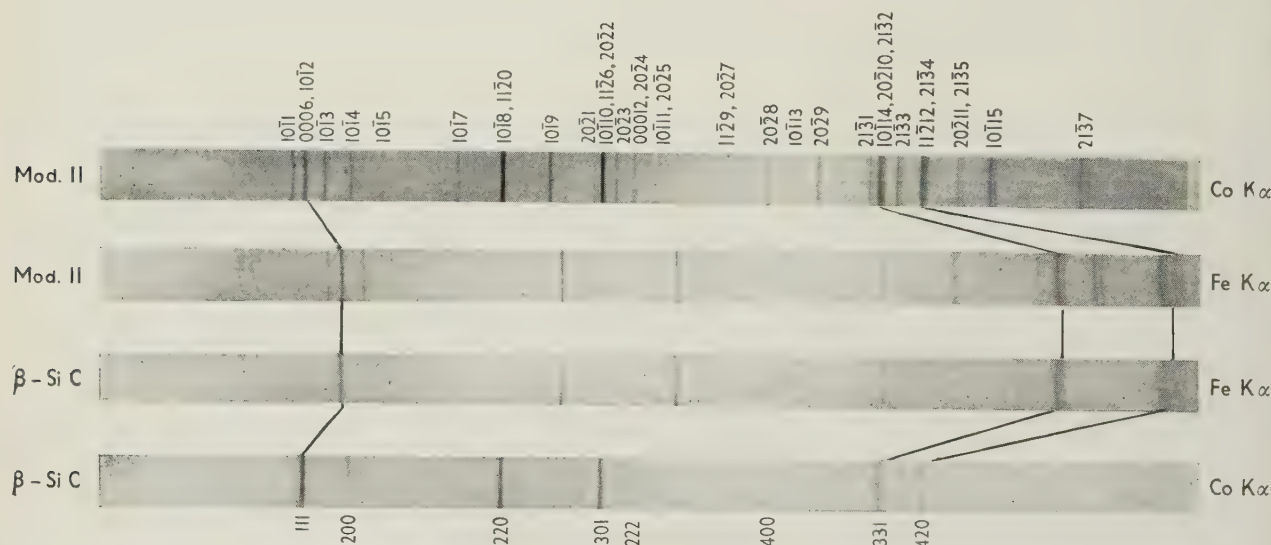


Fig. 3. Powder photographs of various forms of silicon carbide taken with Co  $K\alpha$  and Fe  $K\alpha$  radiation revealing the deviation from ideal axial ratio. The doublet in the cobalt radiation photograph corresponding to the overlapping  $21\bar{3}4$  and  $11\bar{2}12$  reflexions is clearly resolved with iron radiation into the end group of four lines,  $11\bar{2}12\alpha_1$ ,  $21\bar{3}4\alpha_1$ ,  $11\bar{2}12\alpha_2$  and  $21\bar{3}4\alpha_2$  respectively. Rotating anode tube, exposure 1 h at 80 mA and 40 kV peak. 19 cm diameter Debye-Scherrer camera



eter values corrected to absolute angström units  
ere

$$a = 3.082 \text{ \AA} \quad c = 15.67 \text{ \AA}$$

n the basis of these dimensions and the axial ratio  
relationships between the various modifications given  
Table 1, the cube edge of  $\beta$ -SiC should be

$$a_w = 4.365 \text{ \AA}$$

The agreements between the observed and calculated  
 $\sin^2\theta$  were not very good. It was therefore decided to  
determine the lattice parameters of the laboratory  
product,  $\beta$ -SiC, and of the light green commercial  
sample, modification II. For this purpose Fe K $\alpha$   
radiation was used and the powder patterns which reveal  
the great resolution in the high orders are shown in  
Fig. 3. With  $\beta$ -SiC the 420 reflexion falls very close to  
the knife edges at approximately  $84^\circ$ . The length of the  
cube edge at  $19^\circ\text{C}$  obtained by extrapolating against  
the function  $\frac{1}{2}\left(\frac{\cos^2\theta}{\theta} + \frac{\cos^2\theta}{\sin\theta}\right)$  and correcting for  
refractive index,<sup>(18, 19)</sup> is

$$a_w = 4.3590 \pm 0.0001 \text{ \AA}$$

The theoretically possible lattice parameters for the  
hexagonal modification II obtained from  $a_w$  are:

$$\left. \begin{aligned} a &= \frac{a_w}{\sqrt{2}} = 3.0824 \text{ \AA} \\ c &= 2\sqrt{3} \cdot a_w = 15.1004 \text{ \AA} \end{aligned} \right\} c/a = 2\sqrt{6} = 4.89898$$

By deriving the dimensions of the hexagonal modifi-  
cation directly from the cubic form we expect exact  
overlapping of the cubic and hexagonal lines, e.g.  
2 1  $\bar{3}$  4, 1 1  $\bar{2}$  12 hexagonal and the 420 cubic reflexions  
should overlap exactly; so also should the 1 0  $\bar{1}$  14,  
2 0  $\bar{2}$  10, 2 1  $\bar{3}$  2 hexagonal and 330 cubic. As explained  
above, these reflexions appear doubled. A typical  
photometer curve of the 2 1  $\bar{3}$  1-2 1  $\bar{3}$  2 group of hexagonal  
reflexions obtained by means of an accurate null-  
deflexion instrument<sup>(20)</sup> is shown in Fig. 4, with the  
contour resolved into its component lines.

It was found that the cubic lines fell in the mean  
positions of the highly resolved hexagonal groups.  
This ruled out the possibility of a mixture of cubic and  
hexagonal modifications. Calculation of the positions  
of reflexions from modification I (15R) and modifica-  
tion III (4H), ruled out the possibility of a mixture of  
various hexagonal forms. Calculations based on a  
possible and as yet unreported wurtzite structure were  
also made.

The most probable solution was that modification II  
was entirely responsible for the pattern, but that the  
axial ratio was not exactly equal to  $2\sqrt{6}$ . The problem  
was finally solved by deciding from a high resolution  
photograph taken with Fe K $\alpha$  radiation which of the

end doublets was the 2 1  $\bar{3}$  4 and which the 1 1  $\bar{2}$  12  
reflexion, and checking the intensities of all the spectra  
by structure factor calculations.

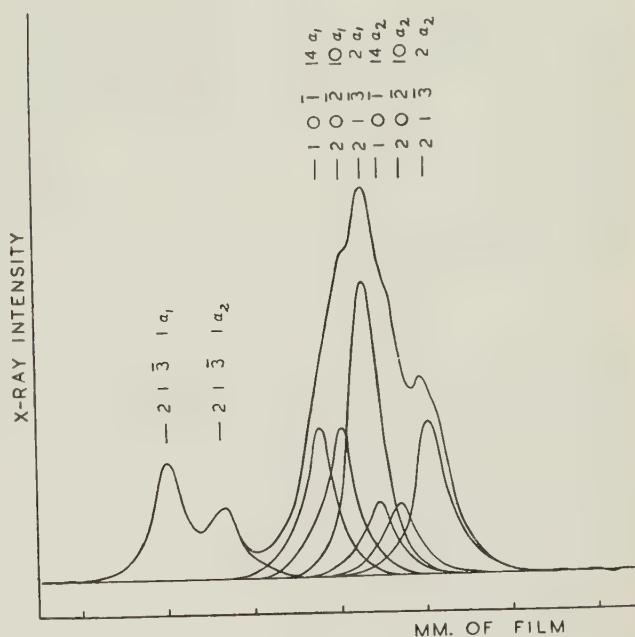


Fig. 4. Photometric resolution of 2 1  $\bar{3}$  1-2 1  $\bar{3}$  2 group of  
reflexions from light green commercial carborundum  
(Modification II). 19.0 cm diameter Debye-Scherrer camera,  
Fe K $\alpha$  radiation

Consider now the end doublets. Their  $\sin^2\theta$  values  
are given by equations of the form

$$\sin^2\theta_{hkl} = \left(\frac{\lambda}{2}\right)^2 \left[ \frac{4(h^2 + hk + k^2)}{3a^2} + \frac{l^2}{c^2} \right]$$

or, writing  $A = [\lambda/\sqrt{(3)a}]^2$ ,  $B = (\lambda/2c)^2$ ,  $s = h^2 + hk + k^2$ ,  
we have:—

$$\sin^2\theta_{hkl} = \sin^2\theta_{hkio} + \sin^2\theta_{ool} = As + Bl^2 \quad (1)$$

Depending on which way we assign the indices to the  
end doublets in the pattern with Fe K $\alpha$  radiation, we  
arrive at two sets of simultaneous equations of the  
type (1). Putting in the measured  $\sin^2\theta$  values, equation  
(1) becomes

$$\left. \begin{aligned} \sin^2\theta_{21\bar{3}4} &= 7A + 16B = 0.98850 \\ \sin^2\theta_{11\bar{2}12} &= 3A + 144B = 0.98690 \end{aligned} \right\} \text{(i)}$$

$$\text{or} \quad \left. \begin{aligned} 7A + 16B &= 0.98690 \\ 3A + 144B &= 0.98850 \end{aligned} \right\} \text{(ii)}$$

Solution (i) yields

$$\begin{aligned} A &= \sin^2\theta_{10\bar{1}0} = 0.13827 \\ B &= \sin^2\theta_{0001} = 0.0041078 \end{aligned}$$

while solution (ii) gives

$$A = \sin^2 \theta_{10\bar{1}0} = 0.131560$$

$$B = \sin^2 \theta_{0001} = 0.0041238$$

The most perfect agreement between calculated and observed  $\sin^2 \theta$ s was obtained, using solution (i). The comparison for Fe  $K\alpha$  radiation after correcting the observed values for drift by the method of Taylor and Floyd<sup>(21)</sup> is made in Table 3, and for Co  $K\alpha$  radiation in Table 4.

The actual dimensions of the unit cell for modification II based on solution (i) are therefore

$$\left. \begin{array}{l} a = 3.0817 \text{ \AA} \\ c = 15.1183 \text{ \AA} \end{array} \right\} c/a = 4.90587$$

These differ from the ideal solution derived from the cubic form:

$$\left. \begin{array}{l} a = 3.0824 \text{ \AA} \\ c = 15.1004 \text{ \AA} \end{array} \right\} c/a = 2\sqrt{6} = 4.89898$$

The lattice parameters of modification II thus deviate quite appreciably from the ideal values based upon the cubic variety. The variations found correspond to a compression of the elementary tetrahedra in the  $a$  direction and an expansion in the  $c$  direction, with corresponding changes in bond angle. The molecular volume, however, remains almost exactly the same. From 19 cm diameter camera powder photographs with Fe  $K\alpha$  radiation, it would appear that the tetrahedra in all the hexagonal modifications undergo exactly the

Table 3. Commercial hexagonal carborundum, Mod. II

$\sin^2 \theta$  values for Fe  $K\alpha$  radiation

Indices	$\sin^2 \theta$ calculated	$\sin^2 \theta$ observed (drift corrected)	$\sin^2 \theta$ for cubic SiC	Indices for cubic lines
2 1 $\bar{3}$ 4	0.98850	0.98850	0.98670	420
1 1 $\bar{2}$ 12	0.98690	0.98690		
2 1 $\bar{3}$ 3	0.95975	0.95972		
$\left\{ \begin{array}{l} 2 1 \bar{3} 2 \\ 2 0 \bar{2} 10 \\ 1 0 \bar{1} 14 \end{array} \right.$	$\left\{ \begin{array}{l} 0.93922 \\ 0.93802 \\ 0.93682 \end{array} \right.$	$\left\{ \begin{array}{l} 0.93914 \\ 0.93802 \\ 0.93674 \end{array} \right.$	0.93821	331
2 1 $\bar{3}$ 1	0.9269	0.9267		
2 0 $\bar{2}$ 9	0.8600	0.8599		
1 0 $\bar{1}$ 13	0.8259	—		
2 0 $\bar{2}$ 8	0.7902	0.7893	0.7901	400
2 0 $\bar{2}$ 7	0.7286	—		
1 1 $\bar{2}$ 9	0.7282	—		
2 0 $\bar{2}$ 5	0.6300	—		
1 0 $\bar{1}$ 11	0.6288	—		
$\left\{ \begin{array}{l} 2 0 \bar{2} 4 \\ 0 0 0 12 \end{array} \right.$	$\left\{ \begin{array}{l} 0.5930 \\ 0.5914 \end{array} \right.$	$\left\{ \begin{array}{l} 0.5919 \\ 0.5919 \end{array} \right.$	0.5926	222
2 0 $\bar{2}$ 3	0.5643	0.5641		
$\left\{ \begin{array}{l} 2 0 \bar{2} 2 \\ 1 1 \bar{2} 6 \\ 1 0 \bar{1} 10 \end{array} \right.$	$\left\{ \begin{array}{l} 0.5437 \\ 0.5433 \\ 0.5425 \end{array} \right.$	$\left\{ \begin{array}{l} 0.5430 \\ 0.5430 \end{array} \right.$	0.5432	311
2 0 $\bar{2}$ 1	0.5314	—		
1 0 $\bar{1}$ 9	0.4645	0.4644		
$\left\{ \begin{array}{l} 1 1 \bar{2} 0 \\ 1 0 \bar{1} 8 \end{array} \right.$	$\left\{ \begin{array}{l} 0.3955 \\ 0.3947 \end{array} \right.$	$\left\{ \begin{array}{l} 0.3948 \\ 0.3948 \end{array} \right.$	0.3950	220
1 0 $\bar{1}$ 7	0.3331	—		
1 0 $\bar{1}$ 5	0.2345	—		
1 0 $\bar{1}$ 4	0.1975	0.1976	0.1975	200
1 0 $\bar{1}$ 3	0.1688	1.1689		
$\left\{ \begin{array}{l} 0 0 0 6 \\ 1 0 \bar{1} 2 \end{array} \right.$	$\left\{ \begin{array}{l} 0.1497 \\ 0.1483 \end{array} \right.$	$\left\{ \begin{array}{l} 0.1482 \\ 0.1482 \end{array} \right.$	0.1481	111
1 0 $\bar{1}$ 1	0.1359	0.1361		

Lines with  $\sin^2 \theta$  values greater than 0.7 were clearly resolved into  $K\alpha_1$  and  $K\alpha_2$  components, but for simplicity only  $\alpha$  values are quoted.

Table 4. Commercial hexagonal carborundum Co  $K\alpha$  radiation

Hexagonal indices	Calc. int.	Obs. int.	Calc. $\sin^2 \theta$	Obs. $\sin^2 \theta$ (drift corrected)	$\sin^2 \theta$ cubic form	Cubic indices
2 1 $\bar{3}$ 7	77	72	0.95982	0.95982		
1 0 $\bar{1}$ 15	95	95	0.9016	0.9015		
$\left\{ \begin{array}{l} 2 1 \bar{3} 5 \\ 2 0 \bar{2} 11 \end{array} \right.$	$\left\{ \begin{array}{l} 28 \\ 28 \end{array} \right.$	$\left\{ \begin{array}{l} 28 \\ 28 \end{array} \right.$	$\left\{ \begin{array}{l} 0.8757 \\ 0.8746 \end{array} \right.$	$\left\{ \begin{array}{l} 0.8750 \\ 0.8750 \end{array} \right.$		
$\left\{ \begin{array}{l} 2 1 \bar{3} 4 \\ 1 1 \bar{2} 12 \end{array} \right.$	$\left\{ \begin{array}{l} 112 \\ 112 \end{array} \right.$	$\left\{ \begin{array}{l} 136 \\ 136 \end{array} \right.$	$\left\{ \begin{array}{l} 0.8441 \\ 0.8427 \end{array} \right.$	$\left\{ \begin{array}{l} 0.8438 \\ 0.8433 \end{array} \right.$	0.8433	420
2 1 $\bar{3}$ 3	63	72	0.8195	0.8193		
$\left\{ \begin{array}{l} 2 1 \bar{3} 2 \\ 2 0 \bar{2} 10 \\ 1 0 \bar{1} 14 \end{array} \right.$	$\left\{ \begin{array}{l} 146 \\ 146 \\ 146 \end{array} \right.$	$\left\{ \begin{array}{l} 164 \\ 164 \\ 164 \end{array} \right.$	$\left\{ \begin{array}{l} 0.8020 \\ 0.8010 \\ 0.8000 \end{array} \right.$	$\left\{ \begin{array}{l} 0.8008 \\ 0.8008 \\ 0.7995 \end{array} \right.$	0.8011	331
2 1 $\bar{3}$ 1	28	35	0.7915	0.7913		
2 0 $\bar{2}$ 9	58	60	0.7343	0.7341		
1 0 $\bar{1}$ 13	6	7	0.7053	0.7050		
2 0 $\bar{2}$ 8	46	45	0.6747	0.6746	0.6746	400
$\left\{ \begin{array}{l} 2 0 \bar{2} 7 \\ 1 1 \bar{2} 9 \end{array} \right.$	$\left\{ \begin{array}{l} 14 \\ 14 \end{array} \right.$	$\left\{ \begin{array}{l} 10 \\ 10 \end{array} \right.$	$\left\{ \begin{array}{l} 0.6221 \\ 0.6218 \end{array} \right.$	$\left\{ \begin{array}{l} 0.6220 \\ 0.6220 \end{array} \right.$		
$\left\{ \begin{array}{l} 2 0 \bar{2} 5 \\ 1 0 \bar{1} 11 \end{array} \right.$	$\left\{ \begin{array}{l} 14 \\ 14 \end{array} \right.$	$\left\{ \begin{array}{l} 12 \\ 12 \end{array} \right.$	$\left\{ \begin{array}{l} 0.5380 \\ 0.5369 \end{array} \right.$	$\left\{ \begin{array}{l} 0.5372 \\ 0.5372 \end{array} \right.$		
$\left\{ \begin{array}{l} 2 0 \bar{2} 4 \\ 0 0 0 12 \end{array} \right.$	$\left\{ \begin{array}{l} 27 \\ 27 \end{array} \right.$	$\left\{ \begin{array}{l} 25 \\ 25 \end{array} \right.$	$\left\{ \begin{array}{l} 0.5064 \\ 0.5050 \end{array} \right.$	$\left\{ \begin{array}{l} 0.5058 \\ 0.5058 \end{array} \right.$	0.5060	222
2 0 $\bar{2}$ 3	31	23	0.4818	0.4819		
$\left\{ \begin{array}{l} 2 0 \bar{2} 2 \\ 1 1 \bar{2} 6 \\ 1 0 \bar{1} 10 \end{array} \right.$	$\left\{ \begin{array}{l} 211 \\ 211 \\ 211 \end{array} \right.$	$\left\{ \begin{array}{l} 187 \\ 187 \\ 187 \end{array} \right.$	$\left\{ \begin{array}{l} 0.4643 \\ 0.4640 \\ 0.4638 \end{array} \right.$	$\left\{ \begin{array}{l} 0.4642 \\ 0.4642 \\ 0.4638 \end{array} \right.$	0.4638	311
2 0 $\bar{2}$ 1	17	12	0.4538	0.4540		
1 0 $\bar{1}$ 9	78	55	0.3966	0.3960		
$\left\{ \begin{array}{l} 1 1 \bar{2} 0 \\ 1 0 \bar{1} 8 \end{array} \right.$	$\left\{ \begin{array}{l} 210 \\ 210 \end{array} \right.$	$\left\{ \begin{array}{l} 190 \\ 190 \end{array} \right.$	$\left\{ \begin{array}{l} 0.3377 \\ 0.3377 \end{array} \right.$	$\left\{ \begin{array}{l} 0.3381 \\ 0.3381 \end{array} \right.$	0.3373	220
1 0 $\bar{1}$ 7	28	20	0.2844	0.2843		
1 0 $\bar{1}$ 5	19	17	0.2003	0.2001		
1 0 $\bar{1}$ 4	48	33	0.1687	0.1685	0.1687	200
1 0 $\bar{1}$ 3	95	87	0.1441	0.1438		
$\left\{ \begin{array}{l} 1 0 \bar{1} 2 \\ 0 0 0 6 \end{array} \right.$	$\left\{ \begin{array}{l} 231 \\ 231 \end{array} \right.$	$\left\{ \begin{array}{l} 242 \\ 242 \end{array} \right.$	$\left\{ \begin{array}{l} 0.1266 \\ 0.1263 \end{array} \right.$	$\left\{ \begin{array}{l} 0.1260 \\ 0.1260 \end{array} \right.$	0.1265	111
1 0 $\bar{1}$ 1	74	83	0.1161	0.1151		

Lines with  $\sin^2 \theta$  values greater than 0.3 were clearly resolved into  $K\alpha_1$  and  $K\alpha_2$  components, but for simplicity only  $K\alpha$  values are quoted.



me degree of distortion. It follows, then, that the parameters published by Thibault and Ramsdell and reproduced in Table 1 require revision. The  $a$ -parameters would then be 3.0817 Å, and the  $c$ -parameters a multiple of 15.1183 Å.

The intensities of the lines were computed on the basis of Ott's structure in the usual manner, employing the formula

$$I = \text{const. } A p e^{-2B(\sin^2 \theta)/\lambda^2} \frac{1 + \cos^2 2\theta}{\sin^2 \theta \cos \theta} [F(hkl)]^2$$

where

$A$  is the absorption factor

$p$  the number of co-operating planes

$e^{-2B(\sin^2 \theta)/\lambda^2}$  the Debye-Waller temperature factor

$\frac{1 + \cos^2 2\theta}{\sin^2 \theta \cos \theta}$  the angular factor

and  $F(hkl)$  the structure factor.

The co-ordinates of the atoms are

$$\text{C} = 000, 00\frac{1}{6}, \frac{1}{3}\frac{2}{3}\frac{1}{6}, \frac{1}{3}\frac{2}{3}\frac{5}{6}, \frac{2}{3}\frac{1}{3}\frac{2}{6}, \frac{2}{3}\frac{1}{3}\frac{4}{6}$$

$$\text{Si} = 00u, 00\frac{1}{6} + u, \frac{1}{3}\frac{2}{3}\frac{1}{6} + u, \frac{1}{3}\frac{2}{3}\frac{5}{6} + u, \frac{2}{3}\frac{1}{3}\frac{2}{6} + u, \frac{2}{3}\frac{1}{3}\frac{4}{6} + u; u = \frac{1}{8}$$

and the space group is  $C_{6v}^4 - C6mc$ .

Excellent agreement with the intensities obtained photometrically confirms the structure to be identical with Ott's modification II. These data are reproduced in Table 4. As no marked extinction effects were to be observed, it seems as if the crystallites of finely ground SiC are essentially mosaic in character.

#### THE DENSITY OF CARBORUNDUM

Density measurements by various authors reveal considerable discrepancies. Values ranging from 3.10 to 3.30 are quoted.<sup>(22)</sup> Perhaps the most accurate determinations have been made by O. Weigel<sup>(23)</sup> who gives the value 3.2104 at 25.7° C and 3.2003 at 114° C. We have calculated the true densities of the cubic form and the hexagonal modification II from the lattice parameters and obtained the following values:—

$$\text{Cubic form, } \beta\text{-SiC} = 3.210$$

$$\text{Hexagonal form, modification II} = 3.208$$

The reduction of symmetry thus leads to a decrease of density equal to 0.063%, which is not nearly enough to account for the wide variations quoted above.

#### ACKNOWLEDGMENTS

The authors would like to take this opportunity of thanking Professor H. L. Riley for his very helpful

interest in the work when it was commenced at King's College, Newcastle-upon-Tyne. They also wish to thank the Carborundum Co. and the Universal Grinding Wheel Co., of Stafford, for the wide variety of specimens which were so kindly put at their disposal, and The English Electric Co. Ltd., Stafford, where X-ray diffraction patterns were taken with the aid of the Peristron rotating anode X-ray tube.

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# Soft Soda Glass to Transmit Bactericidal Radiation

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Small amounts of iron, titanium, sulphur and iodine have a marked effect on the transmission of short wavelength ultra-violet light. Provided due care is taken to keep these impurities to a minimum and to melt under suitable (reducing) conditions, soft glasses of a variety of compositions can be made having excellent transmission at 2 537 Å.

There are many applications for glasses transmitting short ultra-violet radiation, and, in particular, the resonance wavelength (2 537 Å) of the low pressure mercury discharge. They may be used, for example, in ultra-violet microscopy, and in the manufacture of low pressure mercury discharge lamps producing bactericidal radiation (so-called germicidal lamps). Previously the only glasses efficiently transmitting 2 537 Å were low expansion "hard" glasses\*; such glasses and their seals to metal are expensive, a disadvantage in quantity production of lamps and other vacuum devices.

It was aimed to produce 2 537 Å transmitting soft glasses using the available raw materials and melting conditions, which meant that the glass would contain about 0.01% of iron oxide as impurity, this being much lower than the iron oxide content of most commercial glasses, but the minimum which it was considered could be achieved without more expensive special conditions. It had long been considered impossible, however, to make such glasses. Thus D. Starkie and W. E. S. Turner† found that a soda-lime-silica glass, of percentage composition  $\text{SiO}_2$  75,  $\text{CaO}$  10,  $\text{Na}_2\text{O}$  15, and containing 0.01% iron oxide in the reduced (ferrous) state, had the transmission curve shown in Fig. 1, which shows complete absorption at about 2 650 Å; the same glass, with 0.01% iron oxide in the oxidized (ferric) condition, cut off completely at 2 675 Å.

Starkie and Turner had emphasized the importance of ensuring that all the iron was present in the reduced condition. We have found that the addition of small amounts (about 0.03%) of aluminium or silicon powder to the batch gives excellent transmission as shown by the transmission measurements also recorded in Fig. 1. A glass of composition  $\text{SiO}_2$  65,  $\text{B}_2\text{O}_3$  2,  $\text{Na}_2\text{O}$  5.5,  $\text{K}_2\text{O}$  9.5,  $\text{BaO}$  18, and containing 0.011% of iron oxide, expressed as  $\text{Fe}_2\text{O}_3$ , when melted with the addition of 0.035% of aluminium to the batch, has the transmission indicated by curve 4 in Fig. 1; the same glass containing 0.010% iron oxide, expressed as  $\text{Fe}_2\text{O}_3$ , melted without aluminium addition, but otherwise under the same conditions, gives the relatively poor transmission of curve 2 in the same figure.

The barium oxide containing glasses of curves 2 and 4

differ markedly in composition from the lime glass of Starkie and Turner. The use of 0.035% aluminium in melting the latter glass, however, again produced excellent transmission (curve J of Fig. 1), a result much superior to that obtained by Starkie and Turner. It would appear that the base glass composition has a relatively small effect on the transmission properties, the major factor being the state of oxidation of the trace of iron present as an impurity. The inferior results of previous workers are probably to be explained mainly by their failure to achieve efficient reduction of the iron to the ferrous state. There is also the possibility, however, that the glasses of previous workers were poorer because they contained harmful impurities other than iron. In this connexion we have found sulphur, iodine and titanium to be particularly harmful as indicated below.

The author's first encounter with sulphur as a harmful

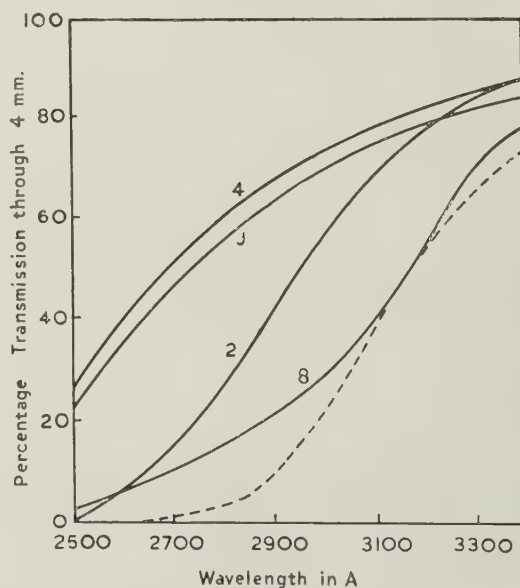


Fig. 1. Transmission of ultra-violet radiation through 4 mm thickness of several glasses. Curve 4: an alkali-barium oxide-silica glass with addition of 0.03% of aluminium to the batch. Curve 2: same glass but with no aluminium addition. Curve 8: same glass using barium carbonate containing a trace of sulphur. Dotted line: soda-lime-silica glass melted by Starkie and Turner. Curve J: same soda-lime-silica composition but melted as described in the text.

\* For example, Pyrex brand glass 9741 and Vycor brand glass No. 791 of the Corning Glass Co. in America. See M. E. Nordberg, *J. Amer. Ceram. Soc.*, **30**, 174 (1947).

† *J. Soc. Glass Tech.*, 1928, **12**, 324 and 1931, **15**, 365.



impurity occurred when attempts were made to melt the glass of curve 4 on a factory scale using materials easily available in large quantities. A slightly yellow glass was obtained, and investigation showed that this was caused by the presence in the barium carbonate of sulphate as an impurity. Thus a melt made under precisely the same conditions as the glass of curve 4, but using the impure barium carbonate, gave the transmission results of curve 8 (Fig. 1).

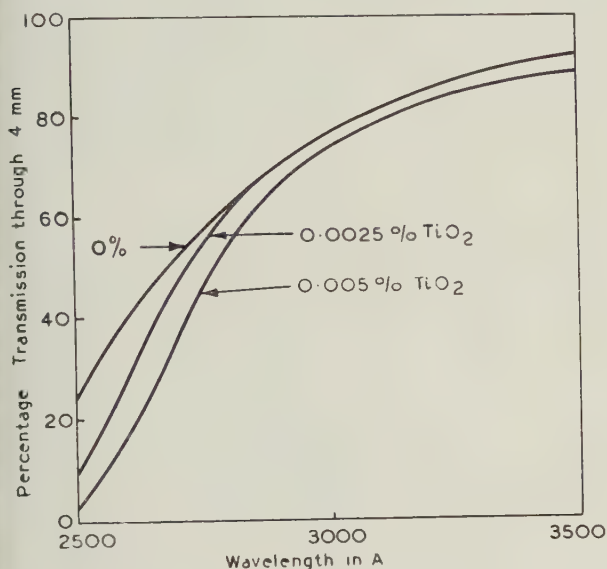


Fig. 2. Effect of titanium on ultra-violet transmission

The second encounter with harmful impurities arose from the presence of titanium in the sand. The melts referred to above had been made with a very good quality American sand, but the best British sand available, however, gave much inferior results in spite of the fact that the iron oxide contents of the melts were sufficiently low. This inferior behaviour of the British sand, at first rather puzzling, received a possible explanation when spectrographic examination revealed that the American sand was free from titanium, whereas the British sand contained titanium as an impurity. A check on the effect of titanium was made by melting glass of curve 4 with additions of titanium oxide to the batch in controlled amounts of 0%, 0.0025%, and 0.005%, reducing conditions again being obtained by the addition of 0.03% of aluminium to the batch. The results of transmission measurements on these glasses are shown in Fig. 2, which clearly shows the harmful effect of titanium.

Evidence has also been obtained that iodine can have a major effect on 2537 Å transmission when added in small quantities to simple borate glasses. A series of melts was made, all of base glass composition Na<sub>2</sub>O 12.4%, B<sub>2</sub>O<sub>3</sub> 87.6%, and with the addition of 0, 0.05, 0.1, 0.3, and 1.0% of potassium iodide to the batch, the melts being made in platinum on a 50 g scale and

using an oxidizing (air) atmosphere. The results are shown in Fig. 3, which clearly shows the marked effect of increasing iodine content. It should be noted that all the glasses were quite colourless in spite of the intense ultra-violet absorption.

To summarize, then, the main conclusion is that small amounts of various impurities, and, in particular, of iron, titanium, sulphur and iodine, have a very marked effect on the transmission of short ultra-violet light, and that a wide variety of glass compositions can have excellent transmission at 2537 Å, provided due care is taken to exclude these impurities. This being the case, it is impossible yet to provide any exact data upon the effect of base glass composition on the ultra-violet absorption. It follows also that the measurement of ultra-violet absorption is of little or no value in the study of the structure of glasses unless extreme care is taken with regard to impurities.

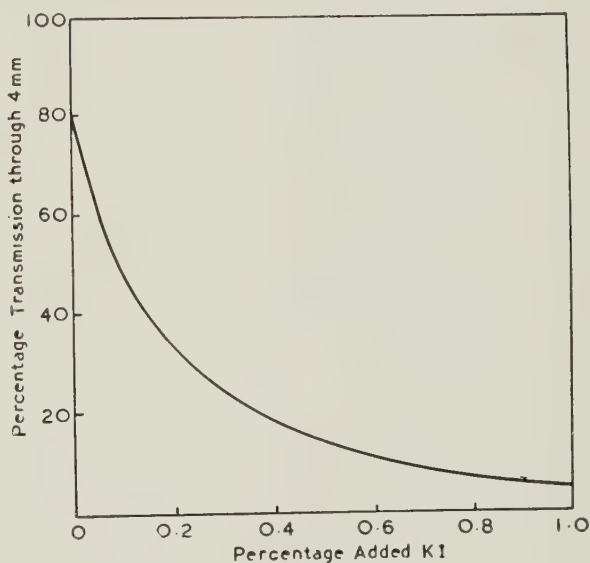


Fig. 3. The effect of potassium iodide additions on the 2537 Å transmission of a simple soda-boric oxide glass

The practical outcome of the work has been the production for the first time on a factory scale of soft soda glasses with excellent 2537 Å transmission. Although no direct observations have been made of the effect on the transmission of these glasses of exposure to ultra-violet radiation, it can be said that germicidal lamps made using the glasses maintain a very satisfactory output of 2537 Å radiation throughout their lives.

#### ACKNOWLEDGMENT

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I also wish to record my appreciation of assistance from many colleagues during the course of the work.

# The Determination of the Principal Stress Differences at a Point in a Three Dimensional Photoelastic Model

By H. T. JESSOP, M.Sc., F.Inst.P., and M. K. WELLS, M.Sc., F.G.S., University College, London

[Paper received 17 March, 1950]

A method is described of finding the directions of the principal axes of stress at any point of a thin slice cut from a "frozen stress" model. The paper includes an account of the method of recording results by a stereographic projection, and a description of the universal tilting microscope stage with a brief investigation into the theory of its use.

## INTRODUCTION

The investigation of stresses in three dimensions at an internal point of a stressed photoelastic model was first made possible by the discovery of the "frozen-stress" property of a number of the plastics used. This property permitted the freezing of a stress-system into a model, after which thin slices could be cut from any part of it without relieving the stresses. Each slice could then be examined in the polariscope and the optical effect produced by the stresses at any point of the model observed. Such an examination, however, does not give a complete solution to the problem, for in order to determine the state of stress at a point we must first find the directions of the principal stresses, and except in two special regions—the free surface of the model or a plane of symmetry—no practicable means of finding these has hitherto been devised. In this paper a method is described by which the directions of the principal stresses and the magnitudes of the principal stress-differences may be found at any point of a "frozen-stress" model. Neither the apparatus (the universal stage and polarizing microscope), nor the method of using it is new, for both have been familiar to mineralogists since the tilting stage was first introduced by E. von Fedorov in 1891. Accounts of the apparatus and the method appear in certain German and American text-books and monographs on mineralogy,\* but to the authors' knowledge no account has hitherto appeared in any British publication.

## GENERAL THEORY OF THE PHOTOELASTIC EFFECT IN THREE DIMENSIONS

The optical properties of a stressed photoelastic material at any point and the effect upon light passing through in any direction may best be represented by the Fresnel's ellipsoid for the material at the point. The property of this ellipsoid is that the length of the radius vector from its centre in any direction is inversely proportional to the velocity of the light wave whose vibrations take place in that direction.

The principal axes  $OA$ ,  $OB$ ,  $OC$  (Fig. 1) of this ellipsoid correspond to the principal axes of stress in the material at the point. If  $a$ ,  $b$  and  $c$  are the velocities of waves

whose vibration directions are respectively  $OA$ ,  $OB$ ,  $OC$ , then  $OA = 1/a$ ,  $OB = 1/b$ ,  $OC = 1/c$ , and the equation of the ellipsoid referred to its principal axes will be

$$a^2x^2 + b^2y^2 + c^2z^2 = 1$$

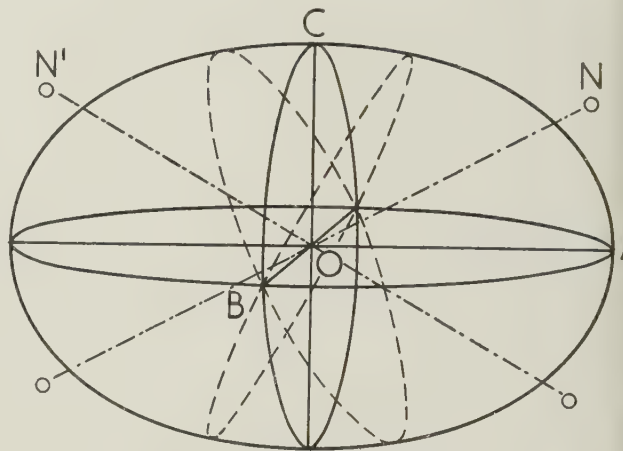


Fig. 1. Diagram of Fresnel's ellipsoid showing principal axes ( $OA > OB > OC$ ), and principal planes. The two circular sections of the ellipsoid, of radius  $OB$ , and the optic axes,  $ON$  and  $ON'$ , are shown by broken lines

If the three axes of the ellipsoid are all of different lengths (say  $OA > OB > OC$ ) there will be two directions in the plane  $OAC$  in which the radius vector is equal to  $OB$ . The sections of the ellipsoid which include these two vectors and  $OB$  will then be circular, and light passing through the medium in the direction of the normal to either one of them will suffer no re-polarization and exhibit no birefringence. These normals,  $ON$  and  $ON'$  (Fig. 1), are called the *optic axes* of the system. They will lie in the plane of the greatest and least vibration axes and be equally inclined to the axis  $OC$ . A system such as this is known as a *biaxial* system.

If two of the principal axes of the ellipsoid are equal, there is only one circular section, viz. that containing these two axes. There is then only one optic axis and this coincides with the third principal axis. The system is then said to be *uniaxial*.

The positions of the optic axes of a crystal are of

\* A. JOHANNSEN: *Manual of Petrographic Method* (New York: 1918), p. 300; M. BEREC: *Mikroskopische Mineralbestimmung mit Hilfe der Universaldrehtischmethoden* (Berlin: 1924); M. REINHARD: *Universal Drehtischmethoden* (Basel: 1931).



importance in mineralogy, and may sometimes be of use in photoelastic applications either in identifying the plane of greatest principal stress-difference, or, in the exceptional case when two optic axes can be located, in determining the directions of the principal axes  $OA$  and  $OC$  which bisect the angles between them.

If now we consider a beam of plane polarized light passing through the medium in any arbitrary direction,  $Om$  (Fig. 2), the beam will in general be split into two polarized waves whose vibration directions will be  $OS$  and  $OT$ , the principal axes of the section of the ellipsoid by the plane of the wave-front.\* If  $V_s$  and  $V_t$  are the

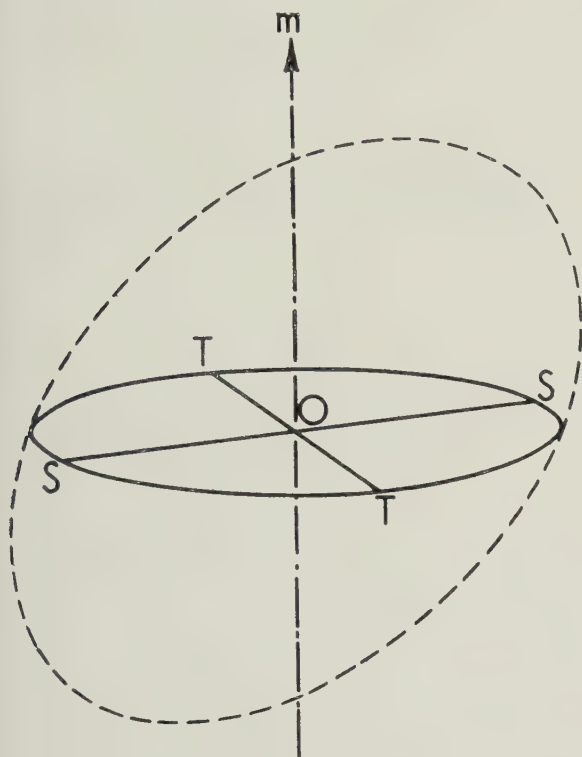


Fig. 2. Section of the ellipsoid by the plane of the wave-front for any ray direction,  $Om$

velocities of these two waves then  $V_s = 1/OS$  and  $V_t = 1/OT$  and  $V_s - V_t$  will be a measure of the birefringence at that point for light travelling in direction  $Om$ . The difference of the normal stresses in directions  $OS$  and  $OT$  is proportional to  $V_s - V_t$ . The problem of finding the principal stress-differences at a point in a slice cut from a frozen-stress model falls into two parts: first, to determine the directions of the axes of the Fresnel's ellipsoid relative to the slice; and second, to orient this slice so that two of these axes lie in the plane of the wave-front, when measurement of the birefringence will give the magnitude of the difference of the corresponding principal stresses.

\* Referred to briefly as "the section of the ellipsoid" in the description which follows.

Fig. 3 shows the microscope with the tilting stage in position. In Fig. 4a the tilting stage is shown with all the essential parts labelled; and in Fig. 4b the latter are indicated in a diagrammatic plan. The stage of the microscope itself can be rotated about the axis  $M$  of the microscope, which is normally kept vertical. This axis is parallel to the light path  $Om$  shown in Figs. 2 and 5.

The central part of the tilting assembly (Fig. 4) consists of a platform which carries the slice to be examined. The platform  $P$  is capable of rotation in its own plane within a graduated ring  $R_1$ , the axis of rotation being referred to as the  $N$ -axis.\* This ring turns about a diametral axis,  $H$ , which is mounted in the outer ring,  $R_2$ , itself capable of tilting about an axis  $K$ , perpendicular to  $H$ . The axis  $K$  is mounted horizontally on the base of the instrument,  $B$ .

(In some models an additional movement is provided by means of which the ring,  $R_2$ , may be rotated relative to the  $K$ -axis, but since this is not required in the procedure which follows, it is omitted from the schematic plan of Fig. 4b.)

In order to eliminate refraction which would otherwise occur with tilting of the slice, the latter is mounted between two segments of a sphere of glass, as shown in Fig. 5. When in position with a glass platform disk,  $D$ , and the slice between them, the segments should form portions of a perfect sphere. The point,  $O$ , to be examined in the slice must lie at the centre of this sphere, and if a suitable immersion fluid is introduced between the contact surfaces, a ray of light,  $Om$ , passing through  $O$  in any direction will suffer negligible refraction. One drop of glycerine (R.I. = 1.47) or cedarwood oil (1.51) placed on each surface is sufficient to ensure good contacts.

\* The initials adopted for the various rotation axes are those used by Reinhard, *Universal Drehtischmethoden*, Basel (1931).

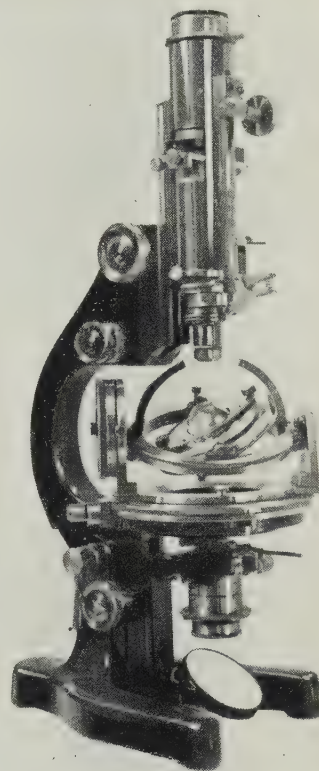


Fig. 3. Microscope and stage

When the stage is set up in the normal working position, the  $K$ -axis is parallel to one of the polarizing directions of the crossed nicols (or polaroid disks) fixed

condition is *the maintenance of extinction when the  $K$ -axis is rotated*, for it is only when an axis is a principal axis of the ellipsoid that it remains a principal axis of all sections of the ellipsoid in which it lies.

It is important to realize that rotations about only two axes are required to place one of the principal axes of the ellipsoid in any desired direction. In the procedure adopted these are the  $H$ - and  $N$ -axes, and it is the co-ordinates of  $N$  and  $H$  which are recorded for graphical representation of the orientation. The functions of the  $K$ -axis and the microscope axis  $M$  are described below.

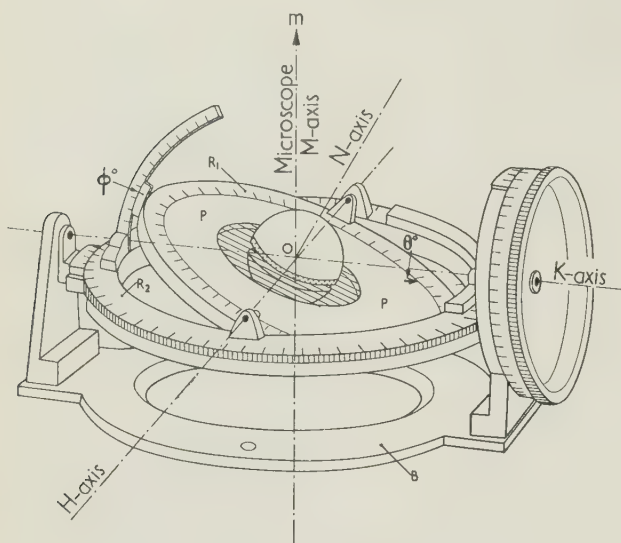


Fig. 4a. Diagram to show the movements of the Universal Stage. The central assembly is shown diagrammatically with the position of the slice stippled, and with the metal support for the upper hemisphere omitted

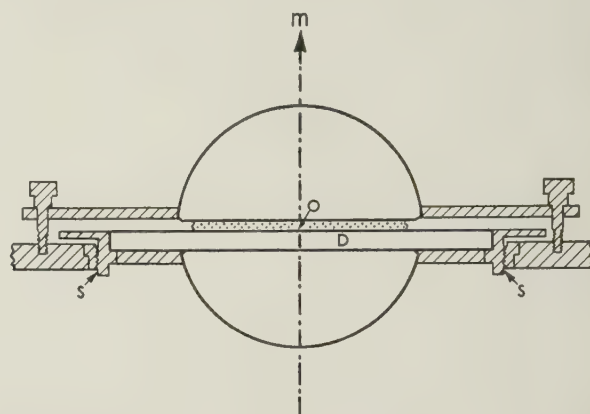


Fig. 5. Assembly on the central platform ( $P$ ) of the Universal Stage. The screw thread,  $SS$ , provides height adjustment for slices of varying thickness

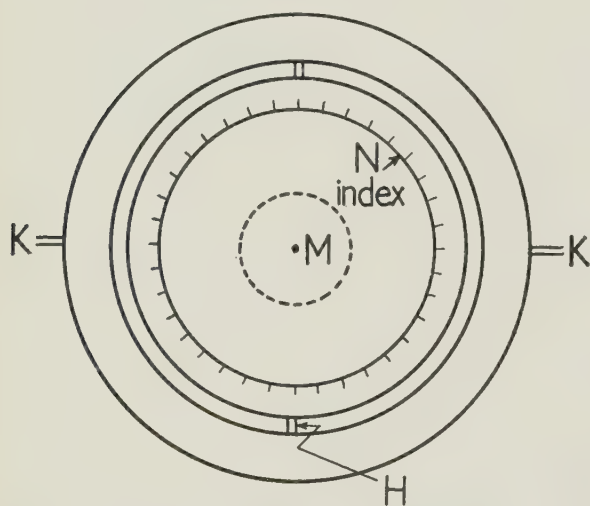


Fig. 4b. Schematic plan of tilting stage

in the microscope. This direction we shall call the east-west direction. It is located with reference to the E-W cross wire fixed in the ocular of all polarizing microscopes. Thus light passing up the microscope axis,  $Om$ , will be extinguished when a principal axis of the section of the ellipsoid (e.g.  $OS$  of Fig. 2) is parallel to the  $K$ -axis. The object of the procedure adopted is to orient the slice (and the ellipsoid at the point observed) in such a way that one of the three principal axes of the ellipsoid itself lies along the  $K$ -axis. The test of this

#### PROCEDURE FOR ORIENTING THE ELLIPSOID TO BRING ONE OF ITS PRINCIPAL AXES INTO THE DIRECTION $KK'$

Having mounted the slice between the glass hemispheres, it is essential to adjust all the movements of the Universal Stage and microscope so that the point of the slice which is being tested remains exactly at the focal point of the microscope during all possible rotations of the stage. (The mechanism of the stage contains facilities for making these adjustments, the details of which are omitted here for the sake of brevity.) Initially the controls are in their zero positions; the  $K$ -axis is parallel to the E-W direction, and the platform and both rings are horizontal. In this position the  $K$ - and  $H$ -scales are both set at zero, and the  $N$ -index of the inner ring is also set at zero. The slice must have some reference grid inscribed upon it, by means of which the desired point of observation,  $O$ , may be located at the centre of the stage (i.e. aligned with the point of intersection of the crosswires of the ocular). It is convenient to start operations with one of the inscribed lines on the slice parallel to the N-S crosswire when the  $N$ -index is in its zero position.

In this setting the orientation of the ellipsoid will be a random one (see Fig. 2) and in general the principal axes of the section in the plane of the wave-front will not be parallel to the polarizing directions of the "nicols,"



so that there will be some degree of illumination at the point under observation.\*

The first step in orientation of the slice is to rotate the platform about the  $N$ -axis until light at the central point of the field is extinguished. In this position one principal

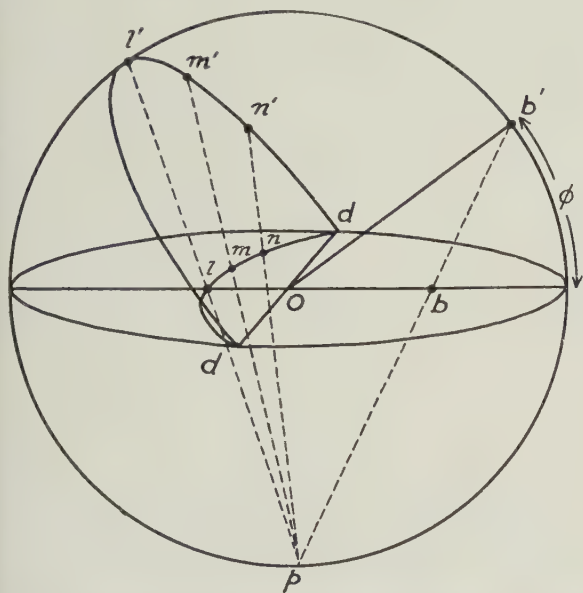


Fig. 6. Stereographic projection of a plane and its normal

axis of the section of the ellipsoid is parallel to  $KK'$ . A test is now made by rotating about the  $K$ -axis in both directions. If it happens that extinction persists during this rotation, then it follows that  $KK'$  coincides with one principal axis of the ellipsoid, and the first part of our problem is solved. If, as will generally happen, light is restored, we proceed with the following sequence of operations:

- (1) Give a small rotation about the  $K$ -axis, in the direction which produces the most rapid restoration of light.
- (2) Tilt the platform about the  $H$ -axis until extinction is restored.
- (3) Give a second small rotation about the  $K$ -axis, but in the opposite direction to the first movement.
- (4) Rotate the platform about the  $N$ -axis to restore extinction again.

This sequence of rotations about axes  $K$ ,  $H$ ,  $K$ ,  $N$  in turn is repeated until rotation about  $K$  fails to restore the light. When this condition is fulfilled the required orientation of the ellipsoid will have been achieved. The co-ordinates of  $H$  and  $N$  (the readings on the respective scales) are recorded and used for plotting the direction of this first principal axis relative to the slice as described below.

\* It should be noted that, due to variations of stress from point to point of the slice, the field will rarely be uniform, and in what follows, all references to extinction or illumination apply only to the region immediately surrounding the central point of the field.

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In the procedure outlined above, the first rotation about  $N$  brought one of the *two* principal axes of the section of the ellipsoid parallel to  $KK'$ . Repetition of the process, but with the *alternative* axis brought to this position will lead to the determination of a second principal axis of the ellipsoid.

Two principal axes of the ellipsoid can always be found in this way with the Universal Stage, and normally the position of the third is found by a graphical construction described in the following section.

## GRAPHICAL PRESENTATION OF THE DATA

Normally the co-ordinates of the three principal axes are plotted on a stereographic projection. The principle of this projection is illustrated by Fig. 6. The circle in this figure represents the outline of an imaginary sphere through the centre of which pass the various planes and axes to be projected. The horizontal diametral plane of the sphere is taken as representing the plane of the slice of material, the point under investigation lying at the centre of the sphere  $O$ . Any axis such as  $Ob'$  meets the sphere at a point, while the plane perpendicular to it meets the sphere in a great circle  $l'm'n'$ . Points and great circles such as these are projected on to the diametral plane by rays to the pole,  $P$ , giving points such as  $b$ , and circular arcs such as  $lmn$ .

In order to simplify the plotting procedure, a von Wulff stereographic net is used as a protractor, Fig. 7.

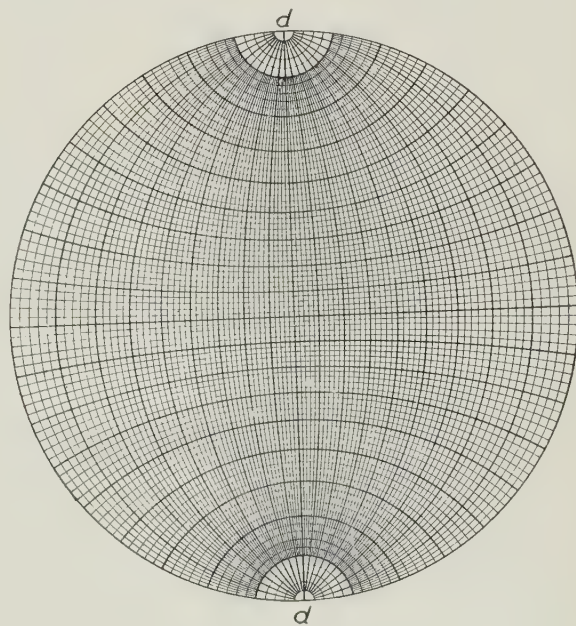


Fig. 7. Stereographic net

This net is mounted on card, and plotting is done on tracing paper which can be rotated over the net, using a drawing pin as a pivot. The outline of the slice is marked on the tracing paper for reference purposes (Fig. 8).

The net has two sets of curves, one the projections of a series of great circles having a common diameter,  $dd$  (Figs. 6 and 7), and the other set the projections of small circles whose centres lie on  $dd$ . The intersections of the

set parallel to the  $K$ -axis of the stage can now be plotted as a point 90 degrees removed from the arc, giving the point  $b$  in Fig. 8.

When the second principal axis is found, its co-ordinates are plotted in exactly the same way as those of the first by rotating the tracing paper according to the value of the  $N$ -reading and selecting a great circle arc to right or left of the diameter,  $dd$ , according to the direction of tilt of  $H$ . At this point there is the first check upon the accuracy of the readings, because one arc should pass through the pole plotted for the other arc. The intersection of the two arcs gives the position of the pole of the third plane, i.e. the position of the third principal axis of the ellipsoid. Again, the accuracy of previous readings can be checked because it is possible to trace in the arc of which this last point is the pole, and this arc should pass through both the previous poles.

#### DETERMINATION OF THE PRINCIPAL STRESS-DIFFERENCES

The directions of the three principal axes having been found, it is a simple matter to read from the stereogram the  $N$  and  $H$  settings necessary to bring any one of the principal planes into the horizontal (for example, in Fig. 9, to bring the plane  $ab$  into the horizontal the  $N$ -setting is  $4^\circ$  and the  $H$ -tilt is  $50^\circ$  to the right). The microscope stage may then be rotated until the principal axes in that plane are inclined at  $45^\circ$  to the vibration directions of the nicols, and the relative retardation is then measured by a compensator inserted into the microscope tube. The length of the light path through the material is calculated from the thickness of the slice and its inclination to the horizontal, and, the fringe value of the material being known, we have all the data needed to calculate the stress-difference.

There are obvious limitations to the amount of information that can be obtained from any one slice. In most photoelastic problems the stresses in the material will vary rapidly from point to point of the model, and to obtain a reasonably accurate determination of the stress conditions at a point we must not only use a very thin slice of the material, but we must also limit the inclination of the slice to the horizontal to about  $45^\circ$  in order that errors due to tilt shall be of the same order as those due to the thickness of the slice. An examination of the stereogram of three mutually perpendicular axes shows that while there can never be more than one principal axis whose inclination to the plane of the slice exceeds  $45^\circ$ , it is possible (as shown in Fig. 9) for the inclinations of all three principal planes to exceed this amount. In this least favourable case the directions of all three axes may be found experimentally, after which two other slices would be needed, each cut parallel to a principal plane, for accurate measurement of the stress-differences. In practice it will usually be found that general considerations of stress-distribution will give some guide as to the most suitable direction in which to cut the first

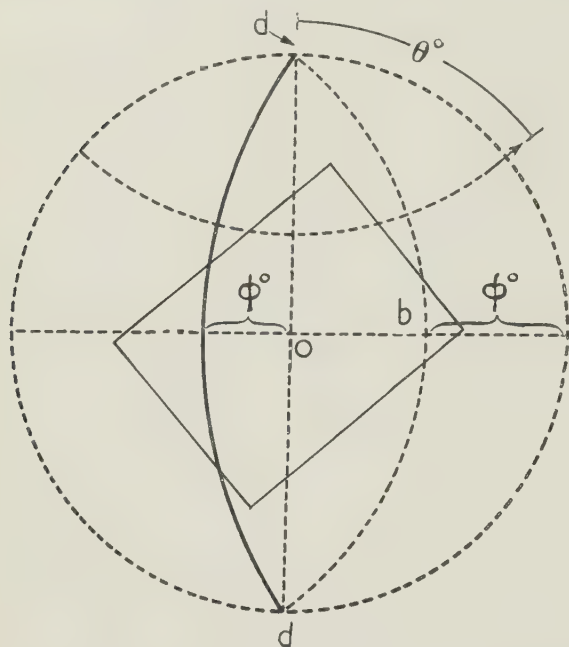


Fig. 8. Diagram to illustrate the use of the stereographic net for plotting the position of a principal plane and axis  $Ob$  normal to it. The relevant arcs of the net are shown by broken lines, and the information drawn on the tracing paper by solid lines

small circle arcs with the perimeter of the net provide a plane protractor for measuring rotations in the plane of the slice ( $N$  rotations). By means of these rotations the diameter,  $dd$ , of the net is made to correspond with the position of the  $H$ -axis relative to the slice, and the tilt about the  $H$ -axis is then recorded by the great circle arcs.

It is convenient to calibrate the perimeter of the net to correspond with the  $N$ -scale of the stage, starting from one end of the diameter,  $dd$ . An index marked on the tracing paper shows the position of the slice relative to the  $N$ -scale.

Suppose that one requires to plot a stereogram of the setting of the stage shown in Fig. 4a, in which the axis of the ellipsoid  $OB$  is lying parallel to  $KK'$ . First rotate the tracing paper clockwise until the index rests on the  $\theta$  degree point on the perimeter of the net (Fig. 8); then choose the appropriate great circle arc to correspond with the tilt about the  $H$ -axis. In this case the platform of the stage was depressed to the right in order to make a principal plane of the ellipsoid vertical: therefore, before tilting, the plane lay  $\phi$  degrees to the left of the vertical. The great circle  $\phi$  degrees to the left of the diameter of the net is therefore traced on to the paper, and corresponds to this principal plane of the ellipsoid. The principal axis of the ellipsoid,  $OB$ , which has been



slices in order that full information may be obtained from two slices only.

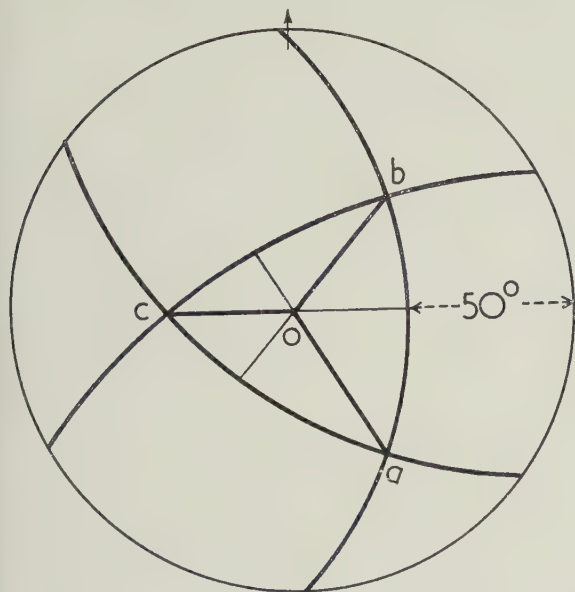


Fig. 9. Completed stereogram of three mutually perpendicular planes

#### THEORY OF THE METHOD

In none of the accounts which the authors have seen has any explanation been given of the way in which the sequence of operations specified results in the alignment of a principal axis in the required direction. The explanation and justification of the procedure follow from a theorem in co-ordinate geometry due to J. W. Archbold. This states that:

If a plane passing through the centre of an ellipsoid be given a small rotation about a principal axis of its section with the ellipsoid, then: (1) the corresponding principal axis of the new section will be greater than the original one if this were a major axis of the section, and less than the original one if this were a minor axis; and (2) the length of the other principal axes of the section will in each case increase or decrease according to the sign of the rotation.

The manner in which the rotations operate is best seen by tracing the path followed by the  $K$ -axis relative to the Fresnel's ellipsoid. Fig. 10 represents one octant of the ellipsoid ( $OA > OB > OC$ ). The dotted curves,  $BD$ ,  $EF$ , etc., are loci of points to which the radius from  $O$  is of constant length,  $BD$  being the central circular section. Let the  $K$ -axis initially cut the ellipsoid in  $K_0$ . Then if  $OK_0$  lies in the segment  $BAD$ , as shown, and is a principal axis of the horizontal section, it must be the major axis, and the tangent to the section at  $K_0$  must coincide with the tangent to the locus  $EF$ .

The sequence of operations may be divided into two groups: first, the pair of rotations about  $OK$  and  $OH$ . Now since the  $K$  rotations are small and in opposite

directions the  $HK$ -plane remains approximately horizontal, and the  $N$ -axis, though progressively tilted by the  $H$ -rotations, remains approximately in the vertical plane through  $OK$ . Thus small rotations about  $ON$  may be considered as the vector sum of rotations about  $OK$  and the vertical axis, while those about  $OH$  are approximately rotations about the second principal axis of the horizontal section. The rotations about the vertical axis will displace  $K$  relative to the ellipsoid along a path  $K1$  or  $K1'$  on the tangent to the locus  $EF$ , while those about the minor axis ( $OH$ ) of the section displace  $K$  in a direction normal to this locus ( $12$  or  $1'2'$ ).

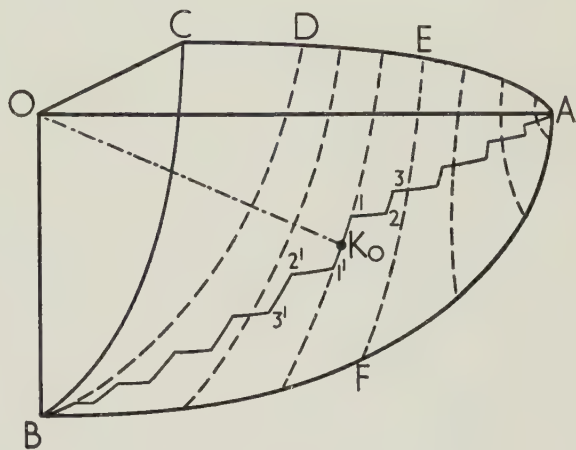


Fig. 10. Path of  $K$ -axis relative to Fresnel's ellipsoid

It may be noted that the  $K$ -rotations do not affect the position of  $K$  relative to the ellipsoid, but only serve to ensure that each  $N$  and  $H$  rotation results in bringing the new principal axis into the direction  $OK$ .

The directions taken by  $K$  in each case are those deduced from Archbold's theorem, viz. for an  $N$ -rotation, always in the direction of increasing major axis, and for an  $H$  rotation, in the direction of increasing major axis for rotations in one sense and decreasing for the other. Thus, starting with the major axis of the section, the method should enable either  $OA$  or  $OB$  to be determined (depending upon the direction of the first  $K$ -rotation) provided both lie within the permissible angle of tilt of the slice.

In a similar manner it may be shown that if the  $K$ -axis initially coincides with the minor axis of the section,  $K_0$  then lying in the segment  $BCD$ , the two alternative paths lead to axes  $OB$  and  $OC$ .

It can be seen from the diagram that the relative magnitudes of the  $N$ - and  $H$ -rotations will be different for different initial positions of  $K$ , and this may sometimes cause failure to find a certain axis at the first attempt. In practice, however, it is found that of the four possible sequences, generally two or three will lead very quickly to well-defined settings, and that the axes and principal planes thus found show agreement within two or three degrees when plotted on the stereogram.

## NOTES AND NEWS

## New Books

**Giant Brains or Machines that Think.** By E. C. BERKELEY. (New York, John Wiley and Sons, Inc.) (London, Chapman and Hall Ltd.) Pp. vii + 270. Price 32s. net.

The main subject of this book is automatic calculating machines, despite anything else which the title might suggest. This is a subject in which there has been considerable progress in the past ten years, and this has stimulated an interest in it which is already wide and is likely to grow as more of these machines are brought to the state of producing results of scientific value. But although there have been articles, and in some cases large volumes, on individual machines and how they are used, there have so far been very few books dealing with the subject as a whole, and a balanced and co-ordinated account of it is to be welcomed.

The title of the present book suggests a lurid and sensational account based more on imagination than on fact, and this suggestion is enforced by the dust-cover which carries a picture of a panel of electronic valves with a cable passing from it to a hole in the forehead of a human face. But the serious reader—if not already deterred by the title and dust-cover—will be relieved to find a sober, unsensational, and well-informed account of the earlier achievements in the field of automatic calculating equipment—punched card equipment, the differential analyser, the Harvard Mark I calculator, the Eniac, and the Bell Telephone Laboratories' relay calculator—and a summary of some of the features of machines still under construction at the time of writing. It also includes an account, the first to be published as far as the reviewer knows, of the Kalin-Burkhart logical truth calculator; though since, as the author explains (with due acknowledgements to Boole) the logical operations involved can be reduced to arithmetical operations on numbers, such a machine can be regarded as a special case of a calculating machine.

The subject is presented almost entirely in terms of individual machines, as is perhaps best for the general reader for whom this book is primarily intended, rather than in terms of general ideas of which individual machines are particular examples. As a result, the reader is not introduced to some of the important ideas only exemplified in machines more recent than those with which this book deals; the most important, however, namely the use of the machine to construct and modify its own operating instructions during a calculation, does receive a bare mention.

There is a valuable classified bibliography, though the entries under "Language; Machines for thinking" would more appropriately come under some heading such as "Calculating machines, general."

The main reservation the reviewer has in recommending this book is concerned with the title and the persistent references in the text to these machines as "mechanical brains" and their operation as "thinking." It is true that the author defines what he means by these terms in the context, and, like Humpty-Dumpty, might reply "When I use a word, it means what I choose it to mean—neither more nor less." But he also, in his preface, invites readers to skip liberally, and many may skip definitions and misunderstand the particular meanings the author attaches to these terms—

and such misunderstanding will be assisted rather than avoided by the dust-cover.

The author seems so obsessed with the idea of these machines "thinking" that he forces biological and psychological terms into his text even when there seems no call for them; for example, he introduces quite unnecessarily the term "mentality" of a machine, and refers to the input elements of a simple machine as "ears" and its output elements as "eyes" that indicate the result by "winking." One may agree that working of this kind makes the presentation more vivid. Whether it makes it more understandable, as is also claimed, seems doubtful: misunderstandable might be the more appropriate word.

He ascribes to the machines the capacity to "think"; but on the other hand he barely mentions the *thought* that the designers have to exert in order to give the machine this capacity, or the *thought* necessary to organize the programmes of operating instructions. But *these* are the steps at which thought is required; once designed and built and furnished with a programme of operating instructions, all the machine can do is to follow these instructions literally and unthinkingly. Over 100 years ago, Lady Lovelace wrote of Babbage's analytical engine that it "has no pretensions whatever to originate anything. It can do whatever *we know how to order it to perform*," and this is still true of the machines so far built.

D. R. HARTREE

**Advanced Plane Geometry.** By C. ZWIKKER. (Amsterdam: North-Holland Publishing Co.) Pp. xii + 299. Price 20s. net.

The author of this book is the Technical Director of the Light Division, N. V. Philips Gloeilampenfabrieken, Eindhoven, who, "led by his own experience in physical engineering," has collected together the most important plane curves and indicated some of their applications in physics. He has, therefore, produced a volume very different from the usual book in English bearing such a title as "advanced plane geometry," particularly as the whole treatment is based on complex numbers.

"In the author's opinion the use of complex quantities in plane geometry has important pedagogic advantages. The student not only learns geometry, but also gets trained in the handling of complex numbers. This method is, moreover, a most appropriate introduction to theoretical electrotechnics, modern acoustics, the theory of complex functions and to operational calculus." However that may be (and we hope the author's optimism is justified), the volume will be read with pleasure by those who teach mathematics to physics and engineering students, for it contains much of value and interest. Anyone convinced of the importance and utility of vector methods will particularly appreciate this book.

The first two chapters are devoted to the representation and properties of complex numbers, with an elementary introduction to conformal and non-conformal transformations. The rest of the book deals with the properties of plane curves, starting with the straight line, the circle and the conic sections, and proceeding successively to involutes, evolutes, caustics, envelopes, and so on, as well as special



rices such as the spirals, the cycloid, epi- and hypocycloids, the cardioid, and many others. The use of complex quantities often results in a very elegant treatment, but occasionally the equations seem clumsy and heavy.

The selected applications in physics are few and are mainly drawn from electrical circuit theory, but the chapter headed "linked curves" provides a simple introduction to the Schwarz-Christoffel transformation and its applications to electrical and hydrodynamical problems.

The book will be of chief interest to mathematicians, but physicists and others will be fascinated by the geometry of many of the curves discussed, and intrigued by the fruitful use of non-cartesian methods. It is not a text-book in the accepted English sense, for there are no examples throughout the book, but it should find a place in school and college libraries. Students who have some acquaintance with the complex variable should find the book stimulating.

The English throughout is clear, though not perfect, and there are certain terms and expressions which might well be amended in a new edition; for instance, such words as *apocissos*, *evolvente*, *trinomium*, and proper names, such as *Cappos*, *Menelaos*, *Ptolemaios*. Again to the English reader the title "*Analytische Dynamik*" for Sir Edmund Whittaker's well-known work will be unusual, as will the term "orthogonal hyperbola," but he will not be misled by the sentence: "We shall meet with more examples of resurrection as we proceed."

J. TOPPING

**Recent Advances in the Physiology of Vision.** By HAMILTON HARTRIDGE, M.A., M.D., M.R.C.P., Sc.D., F.R.S. (London: Messrs. J. and A. Churchill Ltd.) Pp. vii + 401. Price 25s.

The subject matter of this book overlaps the boundaries of physics, physiology and psychology, but there is a strong physical bias in this description and interpretation of the visual process which makes it of direct interest to a physicist. The language throughout is very clear and reasonably free from undefined physiological terms, except for one or two sections which require some proper knowledge of physiology if they are to be properly appreciated. Most of the work described, however, is expressed as some form of optics or spectrophotometry, as, indeed, it must be because the subjective sensation which we call vision can only be examined quantitatively in terms of the physical stimulus.

There are eight chapters, each with about eight short sections with clear titles so that the whole subject is conveniently broken up. The book appears to have been written from notes collected together during the past four or five years, and in several instances there are two sections, widely separated in the book and under different chapter headings, but dealing with much the same aspect of one subject (for example, Eye Movements in Chapter I on p. 64, and in Chapter VII on p. 329). In each section there is a brief and sometimes rather selective sketch of the background; a concise and fair summary of recent papers, published for the most part between 1940 and 1949; comments and criticism of this work and then suggestions for future investigation. The subjects vary widely, but a few section headings may be picked out to illustrate the scope of the book: Light and dark adaptation, Chromatic aberration, Acuity, Retinal functions, Retinal direction effects, Microelectrode technique, Hue discrimination, Theories of colour vision, Retinal pigments, and the Nature of the visual process.

Hartridge's writings are always stimulating and sometimes provocative; in this book he has not hesitated to show clearly where he stands in some matters which are debatable or even controversial. His critical examination of some theories and experimental results is welcome, but some of the rather hypothetical questions which he asks seem to have been included as debating points, perhaps to emphasize that most current theories of the visual process are largely empirical. It must be acknowledged that different experimenters sometimes get different results, and that a convincing proof can only be given by an experimenter who is able to produce either result at will and so to demonstrate the cause of the difference.

This book sets out to survey the whole field of the physiology of vision and it has achieved this object admirably, giving a very useful and fair summary of much of the work published during the past ten or twelve years. It may be recommended to those who wish to improve their general background knowledge and it provides an interesting commentary on some of the more specialized books published during the past three years.

J. G. HOLMES

#### **A Review of Recent Advances in X-Ray Analysis and the Impact of Radiochemistry on Inorganic Chemistry.**

By SIR LAWRENCE BRAGG, O.B.E., M.C., M.A., D.Sc., F.R.S., and H. J. EMELEUS, D.Sc., F.R.I.C., F.R.S. (Cambridge: W. Heffer and Sons Ltd.) Pp. 99. Price 12s. 6d. net.

This book is one of a series containing postgraduate lectures sponsored by the Oil and Colour Chemists' Association and given by eminent scientists on subjects with which their names are especially associated. It is not surprising therefore that, although there are in this particular volume only three lectures from each author on X-ray analysis and the chemistry of radioactive substances, they give the reader a better appreciation of the subjects with less effort than is often possible from the more formal and extensive books. Presuming that the lectures were intended not for an audience specializing in these methods of research, but for the general chemist or young graduate, then they succeed admirably in giving a useful introduction to the theoretical background and depicting the nature of the experimental work. By the well-chosen examples they show clearly the sort of information that can be obtained. Indeed, they should answer many of the questions posed by young chemists contemplating research in one of these branches of science.

Professor Sir Lawrence Bragg elegantly illustrates by the analogy with optical diffraction the essential principles and problems of X-ray crystal structure analysis, and indicates how the problems have been solved for some inorganic structures. In his second lecture the application to metals and alloys is described, and the third is a review of the methods and achievements with organic molecules.

Professor Emeleus's subject lends itself naturally to an historical introduction leading to atomic fission and the properties of the transuranic elements. Experimental methods are then discussed with as much detail as space permits, and finally, the preparation and some uses of tracer elements is described.

These lectures were given in 1947 and 1948, and the delay in publication is particularly to be regretted in a book reviewing recent advances in two subjects in which there is such progressive research in this country.

G. A. JEFFREY

**Rinehart Mathematical Tables, Formulas and Curves.**

By H. D. LARSEN. (London: Chapman and Hall Ltd.) Pp. 264. Price 15s. net.

This is an excellently produced handbook containing 160 pages of tables and 100 pages of miscellaneous mathematical formulae and physical constants.

The main tables are four- and five-figure common logarithms; natural and logarithmic trigonometrical functions (five-figure at intervals of (a) one minute of arc, (b) 0.01 radian, and (c) 0.1 degree); powers, roots and reciprocals; natural logarithms; hyperbolic functions (five-figure, 0.001–3.00 and 3.00–0.05–10.00); exponential functions at a similar interval, giving also the common logarithms of the values; ordinates and areas of the normal probability curve.

Tables giving only four- or five-figure values of functions are necessarily for occasional desk reference but the importance of clarity and accuracy is as great as in tables intended for lengthy computation. These tables have excellent clarity achieved by the choice of a good type face and by good lay-out and the compiler's checking, by calculation and reference to independent sources, should have obtained a high accuracy.

The second part of the book contains an extensive collection of algebraic, geometrical and trigonometrical formulae, five hundred integrals and a number of graphs of standard functions of elementary mathematics. Much of this great mass of facts (such as the three separate statements, under right-angled, equilateral and oblique triangle, that the sum of the three angles of a plane triangle is  $180^\circ$ ) would appear, we hope, to be of doubtful necessity for those for whom the book is intended. Nevertheless this is a satisfying compilation for those who use these "formula omnibuses."

The handbook deserves popularity; its production is well above the average and its price is cheap by the standards we have unfortunately now come to accept. N. CLARKE

**The Acceleration of Particles to High Energies.** (London: The Institute of Physics.) Pp. xii + 58. Price 10s. 6d. net. (11s. 4d. including postage and packing.)

This volume in the Institute's "Physics in Industry" series makes available the essential facts of a rapidly developing division of physical technology, and is based on the papers presented on the last day of the Institute's 1949 Convention held in Buxton, at sessions arranged by its Electronics Group. It is intended for those who are interested in advances in cyclic and linear accelerators for use in applied nucleonic research, in industry and in medicine, but who are not specialized enough in their knowledge to follow the very large number of publications which have appeared in recent years. The sections are: Foreword, by Professor M. L. E. Oliphant; The Cyclotron, by J. Rotblat; Betatrons and Synchrotrons, by F. K. Goward; Electrostatic Generators, by E. S. Shire; Linear Accelerators, by E. S. Shire; and the Discussion on the above papers edited by Mr. V. J. Francis.

**Notes on the Preparation of Contributions to the Institute's Journals and other Publications.** (London: The Institute of Physics.) Price 2s. net.

A new edition of the Notes has been published. The list of contents is as follows: (1) The Institute's Publications.

(2) Types of Papers and Notes accepted for the Journals. (3) Conditions of acceptance of Contributions. (4) Preparation of Text. (5) Form of Script for Submission. (6) Illustrations and Diagrams. (7) Tabular Matter. (8) Mathematics. (9) Numbers, Dates and Units. (10) Correction of Proofs. (11) Authors' Copies and Reprints. (12) Reproduction. (13) Communications. (14) Acknowledgements. (15) Appendix (Spellings and Abbreviations). (16) Short Bibliography.

Authors who are preparing papers are invited to read the booklet before submitting them for publication.

## Journal of Scientific Instruments

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- The Determination of Electron Microscope Magnification. By R. S. Pease.
- An Unbonded Strain Gauge. By S. Baxter and H. A. Vodden.
- Automatic Regulation of Thermionic Emission. By W. Steckelmacher and S. van der Meer.
- The Design of a High-Speed Oscillograph. By H. A. Prime and P. Ravenhill.
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## SPECIAL ARTICLE

## Electrical Conduction in Glass\*

By P. L. KIRBY, M.Sc., A.Inst.P., Research Laboratories, James A. Jobling and Company Limited

The mechanism of electrical conduction in glass is explained in the light of modern theories of its atomic structure, and the electrical migration of "foreign" ions is discussed. The action of the glass electrode is described and the occurrence of the alkaline error is considered from the viewpoint of atomic structure. Compositional variation of conductivity is explained by reference to the valency requirements of the network atoms, and the temperature variation of conductivity is examined. The relative magnitudes of the dielectric absorption current and true conduction current are considered, and the total loss angle calculated at varying temperatures to illustrate the change-over from a dielectric to an ionic conductor. Attention is given to the stabilization effects observed in the annealing range. The observed inflexions in the log resistivity *v.* reciprocal absolute temperature graphs are shown to arise from a different cause from those discovered in the case of ionic crystals. Note is made of certain low-temperature isothermal changes in the conductivity of glass.

## THE VITREOUS STATE

Matter which exists in the vitreous or glassy state has not been subject to the extensive physical analysis which has been applied to other states of matter. Despite the accumulation of considerable knowledge of the physical behaviour of glass, only recently has a small but growing number of workers given attention to the interpretation of the experimental results in an attempt to explain the fundamental nature and atomic structure of glass. Considerable progress in the theoretical study of the vitreous state has been made in the period between the first<sup>(1)</sup> and second<sup>(2)</sup> symposia on the constitution of glass held by the Society of Glass Technology. The divergence of views and the tentative nature of some of the proposals in much of the published work preclude their presentation in a single theory acceptable to all.

At an early stage in the development of theoretical ideas, an attempt to describe the atomic structure of glass was made by W. H. Zachariasen.<sup>(3)</sup> This theory has proved fruitful in so far as the majority of subsequent authors have accepted Zachariasen's hypothesis as a basis for their own work. Further value ensues from the adoption of the fundamental concepts of Zachariasen's theory, because of the ease with which the physical behaviour of glass may be explained in terms of this atomic picture.

Zachariasen regards single component glasses such as vitreous silica ( $\text{SiO}_2$ ), boric oxide ( $\text{B}_2\text{O}_3$ ), and phosphorous pentoxide ( $\text{P}_2\text{O}_5$ ) as consisting of a network of structural units which consist of a highly charged positive ion, surrounded by oxygen ions which are shared with adjacent units. In contrast to similar continuous structures which exist in the crystalline state the network is characterized by a lack of symmetry and a variation of inter-atomic distance about a mean value. In the

case of vitreous silica, the silicon atoms are surrounded by four oxygen atoms lying at the corners of a tetrahedron. The " $\text{SiO}_4$ " structural unit and a two-dimensional picture of the irregular silicon-oxygen network are shown in Fig. 1. These fundamental assertions of Zachariasen's theory have received ample verification from the results of X-ray diffraction analysis. Zachariasen and several other workers have examined

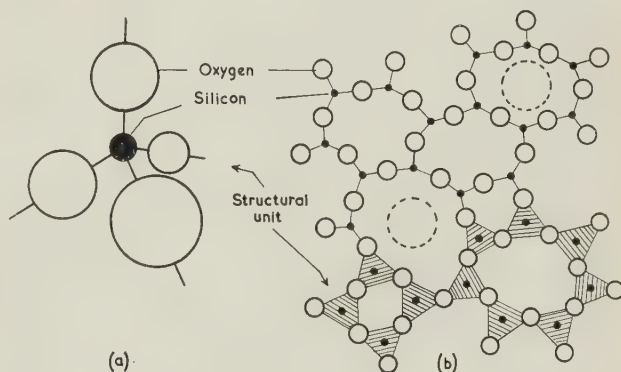


Fig. 1

- (a) The " $\text{SiO}_4$ " structural unit in vitreous silica.  
 (b) Methods of illustrating a two-dimensional picture of the atomic network in vitreous silica ( $\text{SiO}_2$ ). The position of sodium ions in a soda-silica glass is given by the dotted circles.

the valency conditions necessary for the formation of a glass. Analyses of this type have resulted in the division of the constituent positive ions into two classes which have been called "network-forming" ions and "network-modifying" ions, although recent work has shown that certain metals may behave in either manner.<sup>(4)</sup> In the first class are the essential glass-forming atoms such as silicon or boron, which must be present in sufficient quantities in any glass, and whose oxides may themselves exist in the glassy state. Such oxides are

\* Based on a lecture delivered to the North-Eastern Branch of The Institute of Physics on 8 February, 1950.

responsible for the formation of the random continuous network. Other components encountered in the more complex type of glass are the metallic ions which lie in the larger interstices of the network. This addition of, say, the oxide of an alkali metal modifies the structure of the network both by the presence of the interstitial positive ions (indicated by a dotted circle in Fig. 1) and also by the incorporation of the corresponding oxygen ions. Ideally it might be preferable to define the equilibrium position of any ion in terms of the forces acting on it due to its neighbours, and to replace the idea of chemical bonds by the picture of an inter-atomic field of force with the atoms lying at points of minimum potential energy. However, treatments based on the satisfaction of valency requirements have proved of considerable value.<sup>(5)</sup> When the composition of a glass is altered by the addition of a metallic oxide the additional oxygen ions may be absorbed by the network. This happens during the addition of sodium oxide to a boric oxide network where the boron ions may change from a triangular to a tetrahedral co-ordination.<sup>(6)</sup> On the other hand, a stage is reached when, if further metallic oxide is added, the oxygen ions cannot pass into the network in this manner but must lie between adjacent tetrahedral units, acting as a bridging ion. In this case the "structural cohesion" is evidently decreased as suggested by a change in several physical and chemical properties.

With the large variation in charge and radius between a network-forming ion such as silicon and a network-modifying ion such as sodium it is to be expected that the cohesive force maintaining the ion in its equilibrium position in the glassy structure will have different magnitudes in these two cases. The singly charged sodium ion, lying (on account of its size) at a relatively large distance from its neighbours, is able to undergo random diffusion through the medium of the more rigid silicon-oxygen network.

#### MECHANISM OF CONDUCTION IN GLASS

While the majority of glasses exhibit good electrical insulating properties at normal temperatures, it has been known for some time that a rise in temperature is accompanied by a considerable increase in the electrical conductivity. The earliest discoveries of a conduction process in glass, by which is meant a real volume conductivity as distinct from surface leakage effects which were known to Faraday, were made by H. Buff in 1859.<sup>(7)</sup> The early experiments, which are described by J. T. Littleton and G. W. Morey in their monograph,<sup>(8)</sup> while of a simple type, soon brought to light the fundamental nature of the conduction process. E. Warburg<sup>(9)</sup> investigated the passage of an electric current through the walls of a test-tube using internal and external electrodes of mercury. The rapid decrease in current, due to the formation of a high resistance layer in the glass in contact with the mercury anode, was taken to

indicate the occurrence of electrode polarization, which in turn suggested that the process was one of electrolytic conduction. Warburg then found that when a sodium amalgam was used as the anode, no decrease in current was observed. The electrolytic nature of this conduction process has been upheld by the majority of investigators and is readily explained in terms of the aforementioned theories of the atomic arrangement in vitreous matter.

Following Zachariasen's proposals, it is believed that a simple soda-silica glass consists of a random network of silicon and oxygen atoms with the sodium ions lying in the larger voids of the irregular network. It has been suggested that the large size and relatively small charges of the sodium ion endows it with the ability to migrate through the network. This random movement of metallic ions, from one equilibrium position to another through the neighbouring potential barriers of the network, should be capable of study by the application of Boltzmann statistics, not unlike the treatment given by J. Frenkel<sup>(10)</sup> for the diffusion of ions in ionic crystals. The application of an electric field increases the probability of a positively charged ion moving in the direction of the field, by an effective alteration in the height of a potential barrier as shown in Fig. 2. The resulting net migration of the positive ions accounts for the flow of electric charge through the glass.

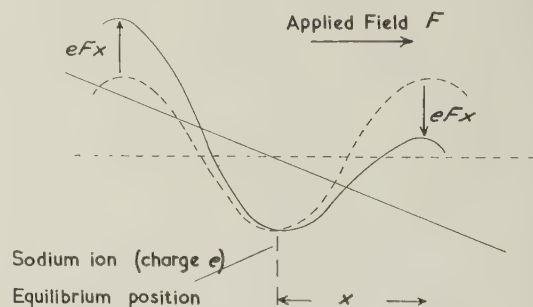


Fig. 2. Change in height of a potential barrier on application of an electric field

----- Original potential diagram.  
 ——— Modified potential diagram.

The author has recently described experiments<sup>(11)</sup> which show that under normal conditions of moderate voltage gradients and slightly elevated temperatures, when glass exhibits appreciable conductivity, Faraday's laws may be verified. An agreement within 0.5% of the accepted value for the electrochemical equivalent of sodium was obtained by measuring the quantity of sodium transported through the glass by a known charge. It is also found that when precautions are taken to ensure that the true conductivity is measured and polarization, dielectric absorption and temperature variation due to Joule's law heating are eliminated, then the conduction process adheres to Ohm's law.

As shown by Warburg's experiment, the unidirectional



flow of current through glass is accompanied by polarization. This is normally associated with the formation of a high-resistance anodic layer with a deficiency of sodium and a resulting liberation of oxygen. If the temperature is sufficiently high the oxygen will come out of solution and small bubbles will appear in the region of the anode. The photograph in Fig. 3 illustrates such an effect in which oxygen bubbles are visible close to the anode while there is no visible change in the glass adjacent to the cathode. When Warburg substituted sodium amalgam in place of the pure mercury anode there was no polarization. In this case the anode

acting on a particular ion. Experimentally it is found that under similar conditions the electrical conductivity of a glass containing sodium will be higher than glass containing equivalent amounts of either potassium or lithium.

#### MIGRATION OF FOREIGN IONS

Even at temperatures at which the viscosity of the glass is high it is possible to bring about the migration of a "foreign" ion, which does not number among the constituents of the glass, into the vitreous network under the influence of an electric field. This is effected by using as the anode a material, such as an amalgam or a fused salt, containing a supply of the positive ions which are to pass into the adjacent glass.

A review of the early experimental work (see monograph by J. T. Littleton and G. W. Morey<sup>(8)</sup>) reveals that in some cases statements made concerning the non-penetration of glass by certain ions have later been contradicted. It is now recognized that many different ions can be made to pass into glass, some, however, in only small quantities. It is always found that penetration is most easily accomplished when the ion concerned is of the same type as one of the metallic ion constituents of the glass. In this respect may be quoted the general conclusion reached by all investigators that extraneous sodium ions may pass through a soda-silica glass, and potassium ions through a potash-silica glass, without either material showing evidence of any physical change. It is also possible to accomplish the transfer of a smaller ion such as lithium into a soda-silica glass, the lithium ions following in the wake of the sodium ions. In such a case the final glass has a white opaque appearance and is liable to fracture spontaneously. Penetration by ions of larger size than those already present in the glass is not readily achieved, the conduction current is small and the final glass is very brittle. The author has found that following the replacement of sodium by silver, with a slightly greater ionic radius, the glass acquires a deep red coloration. The normal soda-silica glass close to the advancing boundary of silver-impregnated glass is found, when examined in a polariscope, to be in a state of tension. The incorporation of a larger ion has evidently caused an increase in the volume of the impregnated glass.

The ease or otherwise with which the ions may migrate through a particular type of glass is shown in Table 1. It is seen that the small platinum ion does not penetrate a normal glass to any extent due, no doubt, to its high charge. An apparent anomaly, in the case of the large ammonium ion, which penetrates glass with facility, is explained by a possible "compressibility" of this complex ion.

As the conduction process in glass is essentially of an ionic nature it is appropriate to compare the phenomenon with the conductivity effects in ionic crystals. Considerable work has been done in recent years on this

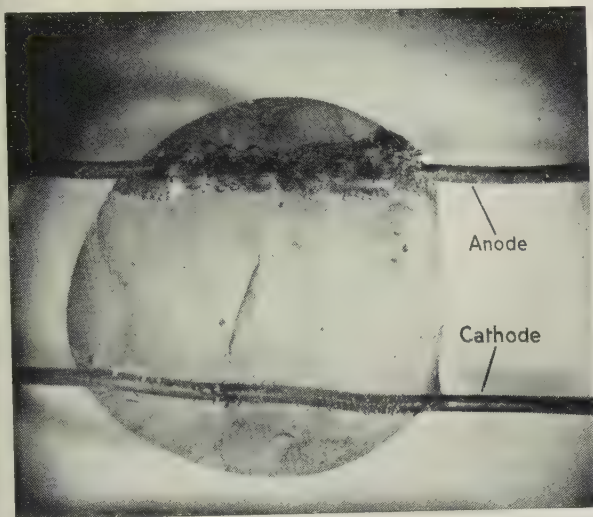


Fig. 3. A bead of soda-silica glass after electrolysis

The anode is surrounded by bubbles of oxygen liberated from the glass. The small bubbles adjacent to the cathode were incidental to the sealing-in process and were present before electrolysis.

behaved as a reservoir for the sodium ions which migrate through the glass. A "replaceable" anode of this type will prevent polarization if it is capable of supplying the type of ion taking part in the conduction process. The absence of any requirement for the cathode to exhibit oxygen replacement illustrates that only the positive metallic ion is responsible for the transit of electric charge through the glass. There is no evidence that negatively charged ions carry any part of the current.

When a glass contains more than one kind of metallic ion there are two important factors which influence the relative mobilities of these ions. A large ionic radius will result in greater separation between the positive ion and the attracting oxygen network, and it is to be expected that the forces of attraction on a sodium ion will be greater than on the larger potassium ion. However, the number of oxygen atoms surrounding the potassium ion is greater than in the case of the sodium ion, that is, the larger ion has a higher co-ordination number. Due to these two opposing factors it is not possible to predict the magnitude of the cohesive forces

latter subject and satisfactory explanations, based on the theories of lattice defects of J. Frenkel, C. Wagner and W. Schottky (survey given in monograph by N. F. Mott and R. W. Gurney<sup>(12)</sup>) have been offered for the observed facts. Such theories are not directly applicable to the very imperfect structure encountered in vitreous matter. The ions responsible for the conduction in glass retain their identity from the random network, whereas the lattice defects in an ionic crystal, whether a hole or an interstitial ion, are associated with the same class of particle from which the ionic lattice is constructed. R. W. Douglas<sup>(13)</sup> has discussed the application to a random glassy network of several recent theories of the liquid state.

Table 1. *Migration of foreign ions into glass*

Ion	Ionic radius (Goldschmidt)	Type of glass	Conduction current	Effect on glass
Li <sup>+</sup>	0.78 Å	Soda	Large	Opaque and brittle
Na <sup>+</sup>	0.98	Soda	Large	None
K <sup>+</sup>	1.33	Soda	Small	Brittle
K <sup>+</sup>	1.33	Potash	Large	None
Cu <sup>+</sup>	0.96	Soda	Large	Coloured dispersion
Ag <sup>+</sup>	1.13	Soda	Large	
Au <sup>+</sup>	1.37	Soda	None	—
Pt <sup>++</sup>	0.52	Soda	None	—
NH <sub>4</sub> <sup>+</sup>	1.59	Soda	Large	None

C. Tubandt<sup>(14)</sup> is responsible for experimental work covering the electrical behaviour of many ionic solids. His work verifies the validity of Faraday's law for several materials to within an accuracy of 1%. During a study of the transport numbers of positive and negative ions, Tubandt has been able to show the existence of electronic conductivity in several cases. He is able to list the salts into three classes, those which under particular conditions exhibit ionic, electronic or mixed types of conductivity.

There is no direct evidence of electronic conductivity in glass, but recent discoveries concerning the emission of secondary electrons from glass<sup>(15)</sup> suggest that it is well not entirely to disregard electronic phenomena when considering its electrical behaviour.

#### THE GLASS ELECTRODE

The glass electrode, used in the determination of the hydrogen ion concentration of solutions, while possessing important practical advantages over a hydrogen electrode, should nevertheless exhibit a pH response similar to the latter. The potential difference measured across a glass electrode system represented by Fig. 4, although made up of a number of contact potentials, will then bear a linear relationship to the logarithm of the concentration of hydrogen ions in the solution "S" under examination. The mechanism of the glass electrode depends on the

ability of the glass to allow the passage of hydrogen ions through the vitreous structure, from a solution whose pH is to be measured on one side, to a standard solution on the other. It has been shown, however, that the thin wall of the glass electrode does not behave merely as a semi-permeable membrane,<sup>(16)</sup> but that each liquid/glass interface acquires a potential difference dependent on the ion concentration in that particular liquid. The equilibrium established between ions in the glass and those in the solution may be treated theoretically in a similar manner to R. W. Gurney's theory of liquid/metal interfacial potential differences. This extension to Gurney's theory has been made by M. Dole.<sup>(17)</sup>

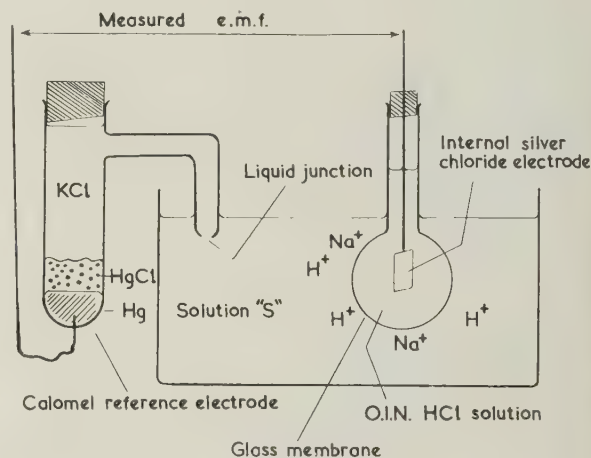


Fig. 4. Simple glass electrode apparatus

When ions other than hydrogen are present in the solution a second interfacial equilibrium may be established, thus developing electrode potentials which will be a measure of the concentration of these other ions. In particular it is found that the small ions of the alkali metals lithium and sodium give rise to lithium and sodium electrode functions at the expense of the desired hydrogen electrode function. The deviation from the perfect hydrogen electrode behaviour becomes apparent when the concentration of hydrogen ions is small compared with that of the alkaline ion responsible for the error, that is, in basic solutions. With the majority of glasses in contact with solutions containing a high proportion of sodium or lithium the transport number governing the transfer of alkali metal ions tends more and more towards unity as the number of hydrogen ions present decreases.

This transmigration of alkali metal ions in preference to hydrogen ions is the cause of the alkaline error associated with the glass electrode, illustrated by the graph in Fig. 5.

It has been known for some time that the alkaline error occurring under particular solution conditions varies according to the composition of the glass used.<sup>(18)</sup> In an attempt to find a glass with negligible alkaline



ror, which would behave as a perfect hydrogen electrode over a wide range of pH values, several investigators have carried out comprehensive studies of the variation of pH response with change in composition. A. Perley has recently reported the results of a study of the electrometric properties of over 500 glasses.<sup>(19)</sup>

The alkaline error of a given electrode may be expected to depend on the sodium content of the glass, but the complete removal of sodium does not eliminate the sodium electrode function in alkaline solutions. Indeed one of the earliest satisfactory pH glasses, Corning 015, was a simple soda-lime-silica glass containing 22% of sodium oxide. Perley has shown<sup>(19)</sup> that the replacement of sodium oxide in a 26% soda, 74% silica glass by an equal amount of lithium oxide only succeeds in decreasing the alkaline error, in a sodium hydroxide solution of fixed pH value, to about one-half of its original value. The residual error in the lithia-silica glass when in contact with the sodium hydroxide solution is ascribed to a base-exchange reaction in which sodium ions from the solution replace the lithium ions in the glass. The newly acquired sodium ions on the glass surface then partake in an interfacial equilibrium endowing the glass with a partial sodium electrode function. If the base-exchange reaction could be prevented by the existence of high cohesive forces within the atomic network of the glass acting on the metallic ions, then there would be no alkaline error. Such conditions may be brought about by the incorporation in the glass of small quantities of certain oxides which may behave in a similar manner to those oxides which exert a stabilizing influence on a glass. If small quantities of aluminium oxide are added

to a silica glass the *trivalent* aluminium ions behave as true glass-forming ions and go into *tetrahedral* coordination. The resulting irregularity involving a region with excess negative charge will provide additional attraction on the metallic ion constituents. Perley suggests that the 3% lanthium oxide content of a lithia-silica pH glass with low alkaline error, behaves as an oxygen modifier in this manner, binding the lithium ions into the network and preventing a superficial base-exchange reaction.

#### THE CONDUCTIVITY OF GLASS

The true volume conductivity, which is an intrinsic property of the glass under examination in contrast to the superficial conductivity which depends on external

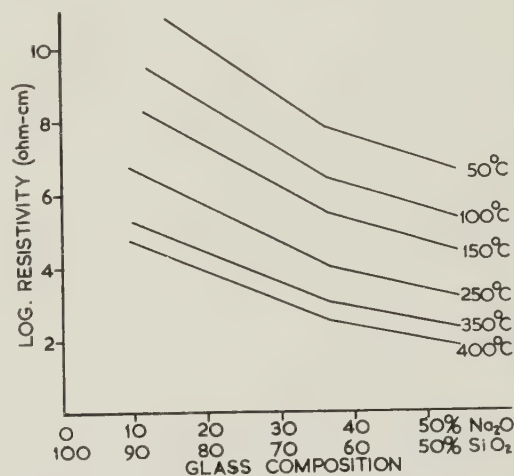


Fig. 6. Variation of resistivity of a soda-silica glass with composition

(After E. Seddon, E. J. Tippet and W. E. S. Turner<sup>(20)</sup>)

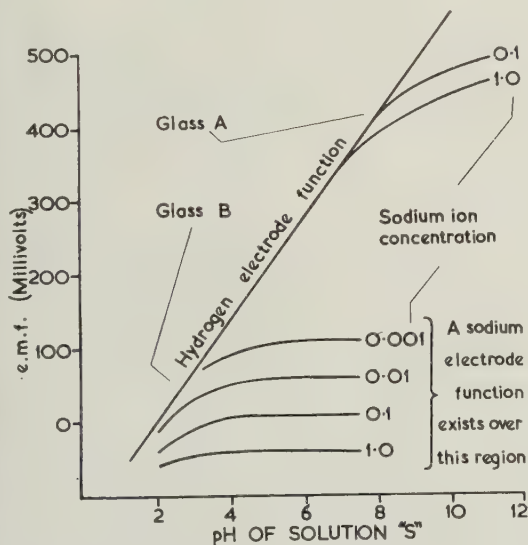


Fig. 5. The pH response of a glass electrode showing the effect of the alkaline error

(After B. Lengyel and E. Blum<sup>(18)</sup>)

"A" would be a more satisfactory glass than "B" for a glass electrode.

conditions, is mainly affected by the composition of the glass and its temperature. While there is only a finite temperature range over which the conductivity of one particular glass may conveniently be measured, there is no practical limit to the extent to which large numbers of oxides may be used to bring about variation of composition.

Confining their attention to a simple two-component glass containing the oxides of silicon and sodium, E. Seddon, E. J. Tippet and W. E. S. Turner<sup>(20)</sup> measured conductivities over a range of temperatures from 50° C to about 500° C. It is interesting to note from their paper that for measurements at any temperature in this range, graphs giving the variation of the logarithm of the resistivity with change in percentage composition (Fig. 6) show inflexions at a composition corresponding closely to that of sodium disilicate ( $\text{Na}_2\text{O} \cdot 2\text{SiO}_2$ ). Although at one time much weight was attached to

evidence of this nature as suggesting the existence of actual chemical compounds in glass, the results are now usually given a different interpretation. It is suggested that during the continual addition of, say, sodium oxide to a pure silicon-oxygen network, a stage is reached at which no further oxygen ions may be assimilated by the network. Further oxygen ions must assume "bridging" positions; a structural change which allows the sodium ions greater mobility. It is possible to imagine that such a transition may well occur at a composition bearing some simple relation to that representing a chemical compound. J. M. Stevels,<sup>(5)</sup> basing his calculations purely on valency considerations, is able to predict the composition in the sodium oxide-silica system at which the structural change occurs; his theory is supported by the appearance of inflexions in the electrical conductivity graphs at the predicted composition.

From the chemical aspect of the constitution of glass it is to be expected that experimental observations of compositional variation of electrical properties will increase our knowledge of the valency co-ordinations of the glass structure. On the other hand, the analysis of physical problems concerning the finer details of the lattice structure, which are not revealed by the average results obtained from X-ray diffraction experiments, may well be furthered by the investigation of the temperature variation of electrical conductivity.

#### TEMPERATURE VARIATION OF ELECTRICAL PROPERTIES

The wide range over which the conductivity of glass varies as it is heated from room temperature to those temperatures used for the melting and working of the commercial types of glass, is responsible for considerable differences in the electrical behaviour at either end of this temperature range. At room temperature the electrical properties are those of a typical insulator with a resistivity as high as  $10^{16}$  ohm-cm. On heating to, say,  $1\,000^\circ\text{C}$ , the majority of glasses exhibit the electrical behaviour of a pyroelectric and the appreciable conductivity observed will continue to increase with further rise in temperature. The resistivity may fall to 10 ohm-cm, in the region of  $1\,400^\circ\text{C}$ . During the measurement of the conductivity of glass it is essential that due appreciation be given to the large possible variation in electrical behaviour. The apparatus used must possess different features at opposite ends of the temperature range to allow accurate measurements to be made under the extreme conditions encountered.

When the glass must be treated as a dielectric material the application of a unidirectional electric field results in a displacement of charge through the medium. Although the exact nature of the polar elements causing this initial current is not known, the current is undoubtedly greater than the final conduction current associated with the movement of sodium ions. From its minimum value when direct currents are used the in-phase current

increases with increasing frequency, an effect which may be ascribed to the appearance of a frequency dependent dielectric absorption current in addition to the normal conduction current.

The gradual transition from insulator to ionic conductor which a glass undergoes on heating may be analysed in terms of common electrical parameters. The simplest equivalent circuit representing the electrical behaviour of a glass slab lying between plane electrodes will consist of a capacitance  $C$ , and a parallel conductance  $G$ . In this simplified treatment these terms, in addition to representing the pure capacity and d.c. conduction, also incorporate parameters representing the dielectric absorption current. The capacity current  $I_C$  and the in-phase conduction current ( $I_D + I_R$ ) are illustrated in Fig. 7(a), where the resulting current vector

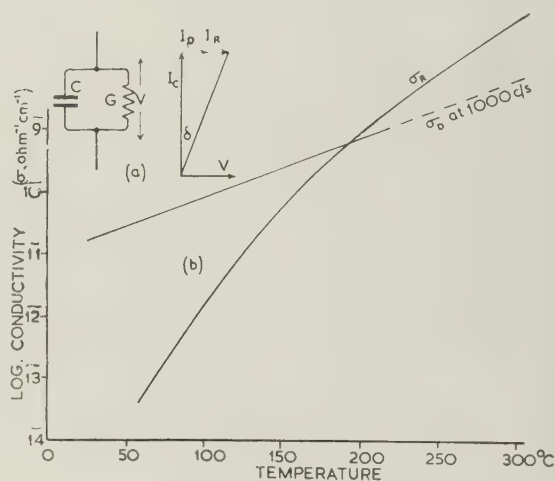


Fig. 7

(a) The equivalent circuit of a slab of glass (dielectric constant,  $k$ ) of thickness  $d$  cm, and area  $A$  cm<sup>2</sup>. An applied alternating e.m.f. of  $V$  volts at a frequency  $f$  c/s, generates currents:

$$I_C = VfkA/2d$$

and

$$(I_D + I_R) = V\sigma_R A/d$$

whence

$$\tan \delta = 2\sigma/fk$$

Expressing both currents in the same units, the loss factor  $k \tan \delta = 9 \times 10^{11} \times 2\sigma/fk$ .

(b) Variation with temperature of the normal d.c. conductivity ( $\sigma_R$ ), and the "absorption conductivity" ( $\sigma_D$ ) at  $1\,000$  c/s, for Pyrex Brand glass.

is defined by the loss angle,  $\tan \delta$ .  $I_R$  is the normal d.c. conduction and  $I_D$  the absorption current which disappears at zero frequency. If the dielectric constant of the material is  $k$ , then the loss factor is  $k \tan \delta$ , which may be expressed in terms of the overall conductivity  $\sigma$ . When self-consistent units are used  $k \tan \delta = 18 \times 10^{11} \sigma/f$ . The conductivity  $\sigma$  may be resolved into two components, the normal d.c. conductivity,  $\sigma_R$ , and the "absorption conductivity,"  $\sigma_D$ . It is generally agreed that the in-phase current at low temperature consists



mainly of the dielectric absorption current, while at high temperatures the true conduction current predominates. To illustrate this change-over in the relative magnitudes of these components the following calculations have been made, based on work done by L. S. McDowell and H. L. Begeman.<sup>(21)</sup> These workers measured the loss angle for Pyrex Brand glass at a frequency of 1 000 c/s, and have corrected their values to eliminate the portion of the in-phase current due to d.c. conduction. Hence in the absence of  $\sigma_R$  the loss factor is given by  $k \tan \delta = 18 \times 10^{11} \sigma_D / f$ , from which it is possible to calculate  $\sigma_D$  over a range of temperatures. The results are shown in Fig. 7(b) together with known values for the d.c. conductivity  $\sigma_R$ , for this glass. It must be remembered that  $\sigma_D$  is not an intrinsic conductivity in so far as it depends on the frequency of the applied alternating e.m.f. In this particular case (1 000 c/s) it is seen that  $\sigma_D = \sigma_R$  at a temperature of about 200°C, and bearing in mind the logarithmic nature of the ordinate scale, it will be seen that at slightly lower temperatures  $\sigma_R$  becomes negligible compared with the absorption conductivity. If the glass is heated above this temperature the normal conduction current predominates.

It is instructive to compare these deductions with certain results due to D. M. Robinson,<sup>(22)</sup> who showed that measurements of the conductivity of Pyrex Brand glass at varying temperatures carried out using currents alternating at 1 000 c/s, merged into the values obtained from d.c., measurements in the region of 200°C, as shown in Fig. 8. All of Robinson's conductivity

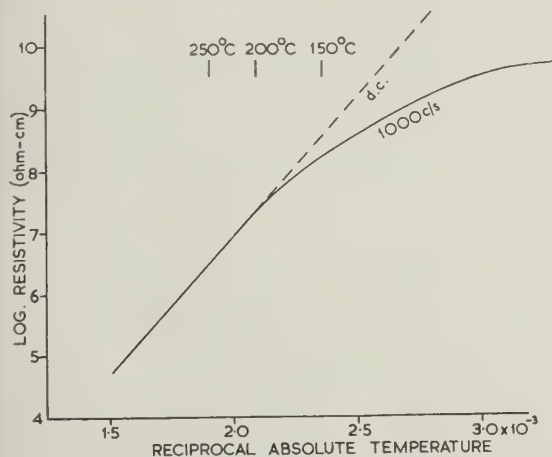


Fig. 8. Temperature variation of resistivity of Pyrex Brand glass using direct current and alternating current at 1 000 c/s.

(After D. M. Robinson<sup>(22)</sup>)

measurements for this glass are higher than those made by the author and used to calculate  $\sigma_R$  in Fig. 7. Nevertheless, his results show that the difference between d.c., and 1 000 c/s, measurements become appreciable due to the occurrence of dielectric absorption currents

at the temperature predicted by the calculation given above.

Further calculations involving the summation of  $\sigma_D$  and  $\sigma_R$  allow the temperature variation of the total loss angle to be predicted. Values are given in Fig. 9 for Pyrex Brand glass at a frequency of 1 000 c/s, and it is seen that between the temperature 200°C and 300°C the material changes its essential electrical nature from an insulator to an ionic conductor.

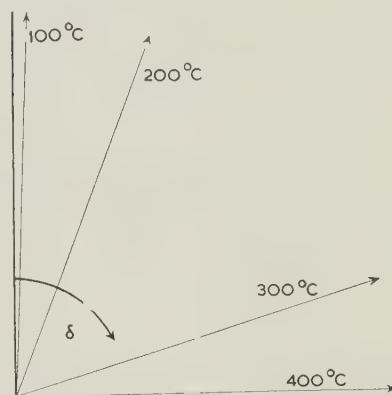


Fig. 9. Temperature variation of loss angle for Pyrex Brand glass at a frequency of 1 000 c/s

Conductivity measurements at low temperatures are liable to error due to the presence of the dielectric absorption component. If direct currents are used time must elapse to allow the full decay of the anomalous charging current before conductivity measurements are made. The bulk movement of sodium ions, giving rise to electrode polarization, may cause further error which can, however, be avoided either by the application of low-frequency alternating currents or by the use of "replaceable" electrodes, thus preventing the formation of a high-resistance anodic layer. If alternating currents are used the frequency should increase with rising temperature to prevent the occurrence of cyclic polarization, but should never be high enough to permit interference from dielectric absorption or the wattless capacity currents. At temperatures above about 500°C, the normal conduction current is greatly in excess of the dielectric currents (even for frequencies as high as 1 Mc/s) and the material may be regarded as an ionic conductor.

#### TEMPERATURE VARIATION OF CONDUCTIVITY

The temperature-resistivity behaviour of any glass may be expressed over limited temperature ranges by the equation proposed by E. Rasch and F. W. Hinrichsen<sup>(23)</sup>

$$\log \rho = A/T + B$$

where  $\rho$  represents the resistivity at a temperature

$T^\circ$  abs., while the constants  $A$  and  $B$  will vary from one glass to another. The existence of certain deviations from this ideal has been known for some time. Measurement of the conductivity of a glass from room temperature up to, say,  $1\,200^\circ\text{C}$  show that there are two temperature regions above and below some temperature in the annealing range, in which a linear relationship exists with different slopes on a log. resistivity versus reciprocal absolute temperature graph. At high temperatures a continual curvature is apparent. The sharp inflexion in the curve in the annealing range has also been detected during the measurement of other temperature-dependent properties of glass. The tendency towards the belief that there existed a definite transition temperature was halted by J. T. Littleton,<sup>(24)</sup> who showed that the temperature at which the discontinuity occurred varied with the rate of change of temperature undergone by the glass during the experiment. This effect is illustrated for decreasing temperature in Fig. 10.

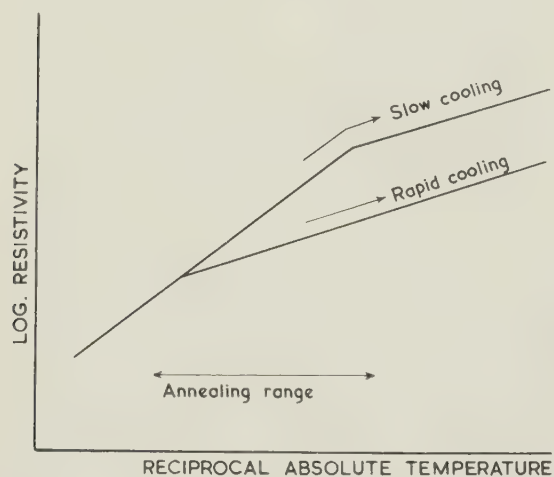


Fig. 10. Change in the temperature of the discontinuity with rate of cooling

It has since become clear that for a glass at any particular temperature there exists a certain equilibrium state, while the rate at which the material attains this fully stabilized or equilibrium condition will increase rapidly with rising temperature. J. T. Littleton and W. L. Wetmore<sup>(25)</sup> investigated this phenomenon by measuring the resistivity of a soda-potash-lead glass, while the author has recently completed a series of measurements of a similar effect found with Pyrex Brand glass.

By imposing on the glass a rapid temperature change there occurs a corresponding change in electrical resistivity. The glass now stabilizes at its new temperature, thereby undergoing an additional resistivity change, as shown in Fig. 11(a). This latter process has been examined and it has been found that the time necessary to achieve stabilization varies from several

days at  $450^\circ\text{C}$  to a few seconds at  $650^\circ\text{C}$ . The variation of resistivity during stabilization, illustrated in Fig. 11(b), during the asymptotic approach to a final equilibrium value, is not capable of exact representation by a simple exponential function. Thus there is no true half-value period. However, so that the order of the time factor during stabilization may be assessed, a mean half-value period, deduced from several parts of the stabilization curve, has been determined. These values were measured at several temperatures in the annealing range and indicate a linear relationship between the logarithm of the mean half-value period and the reciprocal absolute temperature (Fig. 12). Experiments performed on the same glass to ascertain the variation of thermal expansion\* during stabilization give mean half-value periods in close agreement with those obtained from electrical conductivity measurements.

An explanation may be given of the apparent transitional effect observed when resistivity is measured with changing temperature in the annealing range. The

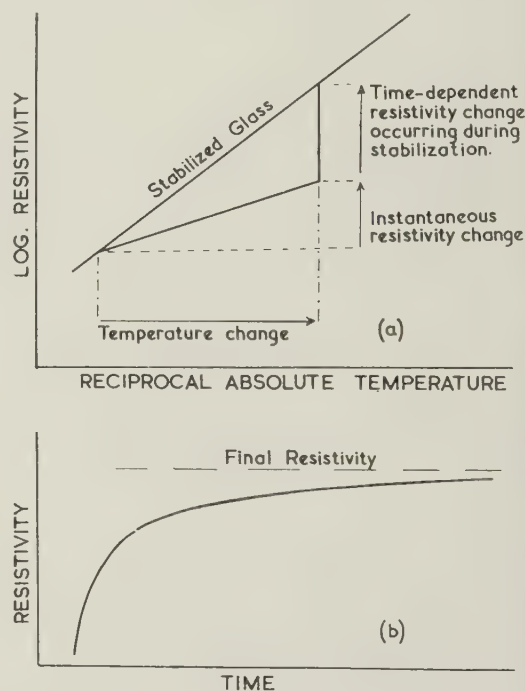


Fig. 11

(a) The initial resistivity change caused by an "instantaneous" temperature change, followed by an isothermal resistivity increase while the glass stabilizes.

(b) Resistivity change during stabilization.

temperature at which a break occurs in the property versus temperature graph will be near to that at which the rate of stabilization attains a similar value to the

\* Private communication concerning unpublished work from Dr. S. M. Cox, of these laboratories.



te at which the temperature is changing. At temperatures above the inflexion the glass is fully stabilized, while at lower temperatures the equilibrium condition is not achieved. The apparent sharpness of the observed discontinuity is due to the large variation of the rate of stabilization over a small temperature range. Rapid cooling through the critical temperature region will result in a final glass with properties indicative of a high-temperature state which has been "frozen" into the material. Chilled glass thus exhibits greater conductivity than a similar well-annealed specimen.

It is apparent that the resistivity-temperature behaviour of a glass in its annealing range bears a superficial relationship to that of an ionic crystal, the resistivities of the latter also showing discontinuities at a particular temperature. The work of W. Lehfeldt<sup>(26)</sup> may be cited in this connexion. Following C. Tubandt's study<sup>(14)</sup> of electrical conduction in ionic crystals, Lehfeldt investigated the effect of temperature variation. He found a transition temperature for several salts ranging from 600°C for sodium chloride to below 0°C for silver bromide. Above the transition temperature it is possible to obtain repeatable results using either single

evidence, nor theoretical reason, to suggest that previous heat treatment will effect the conductivity of an ionic crystal, except in so far as this may alter the state of dispersion of an impurity in the lattice.

#### LOW TEMPERATURE RESULTS

Measurements of the stabilization half-value periods for resistivity changes occurring in Pyrex Brand glass have been carried out at temperatures down to 440°C. Extrapolation to room temperature suggests that the half-value period rises to  $10^{25}$  years. It has been shown, however, that isothermal changes in physical properties of a glass may occur at a much greater rate than that indicated by this extrapolation from the annealing range. E. Seddon<sup>(29)</sup> has measured the changes in resistivity which occurred in chilled and annealed specimens of a glass while held at room temperature. Measurements were taken over a period of three years and showed an increase of 30% in the resistance of the chilled glass and about 16% increase for the annealed specimen. If these results are to be accounted for by the same type of structural change which gives rise to the stabilization effects in the annealing range then the time order of the low temperature results may be due to the relatively high rate of change during the early stage of a stabilization process. It will be noted that extrapolation of the graph in Fig. 12 to the hypothetical state  $1/T^\circ \text{ abs.} = 0$ , shows that the most rapid structural change which can occur will do so in a time similar to the period of an atomic vibration.

#### CONCLUSION

The foregoing survey shows that there is no lack of experimental results concerning the many aspects of electrical conduction in glass. It would now seem appropriate to pursue further investigation in each branch of the subject in the hope of uniting them, together with a theoretical explanation of the electrical behaviour of glass. It is to be expected that at this stage the important recent advances in the development of theoretical ideas will suggest the most profitable lines for future experimental research.

#### ACKNOWLEDGEMENTS

Acknowledgement is made to the Directors of James A. Jobling and Co. Ltd., in whose laboratories some of the work mentioned was performed, for permission to publish this paper. The author also expresses his gratitude to the Research Manager of the above laboratories, Dr. S. M. Cox, and to Mr. J. F. Stirling for their encouragement and helpful comments. Thanks are also due to Dr. W. A. Prowse of the Durham Colleges Science Laboratories for a valued criticism of the section dealing with dielectric absorption.

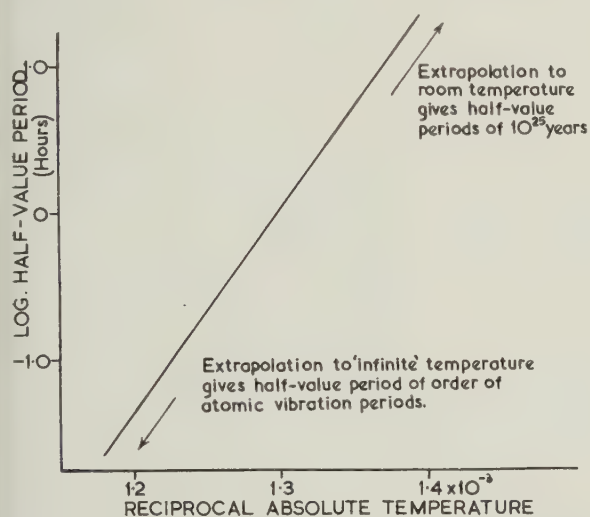


Fig. 12. Mean half-value periods for the stabilization of Pyrex Brand glass in the annealing range

crystals or polycrystalline material. Below the transition temperature the results are greatly dependent on small quantities of impurities in the crystal. E. Koch and C. Wagner<sup>(27)</sup> measured the conductivity of ionic crystals containing varying small quantities of impurities. The resulting defects in the lattice structure cause an increase in the conductivity. In this case, however, although the "foreign" ions are modifying the structure it is, nevertheless, the lattice ions which constitute the current carriers. It is, therefore, not to be expected that conditions are in any real sense analogous to those obtaining in a glass. N. F. Mott and R. W. Gurney<sup>(28)</sup> report that there is no

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## ORIGINAL CONTRIBUTIONS

## The Change of Viscosity of Oils containing High Polymers when Subjected to High Rates of Shear

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[Paper received 22 November, 1949, and in final form 12 May, 1950]

The construction and use of an apparatus for the imposition of rates of shear of at least  $10^7 \text{ secs}^{-1}$  on oils containing high polymers is described. The apparatus is constructed from standard fuel injection equipment with minor modifications. A series of typical results is reported, showing that a substantial decrease in the intrinsic viscosity of the polymer, and consequently in the viscosity of the solution, can occur on application of rates of shear of the above order.

It is by now well known that the addition of a small quantity of oil-soluble high polymeric material to a mineral oil fraction has a profound effect on the temperature dependence of the viscosity of the oil. As the effect is such that the temperature dependence is decreased, high polymers are finding increasing use in the oil industry in applications where oils are subjected to extremes of temperature, as, for example, in the production of lubricating and hydraulic oils for aircraft.

In most applications oils containing high polymers are subjected to rates of shear of a high order, and Morris and Schnurmann<sup>(1,2,3)</sup> have been able to show that both a temporary and a permanent loss of viscosity occur when such rates of shear are applied.

It is considered that when the shear stress is applied to the solution the polymer chains will tend to be

preferentially oriented in the direction of flow of the liquid, causing an apparent or temporary decrease in viscosity. That this decrease in viscosity is temporary is shown by the fact that a viscosity determination at a very low rate of shear, such as obtains in an Ostwald viscometer, after removal of shear stress shows no deviation from that determined before imposition of the stress.

However, if the rate of shear is further increased a point will be reached, dependent upon the nature and chain length of the polymer, when the largest chains present will be ruptured, producing fragments of shorter chain length. When this occurs a permanent decrease in viscosity of the solution is observed, due to the overall reduction in chain length of the polymer. The breakdown of polymer chains into smaller units results in an



erall decrease in viscosity of the solution accompanied an increase in the temperature dependence of viscosity of the solution. It is clear from the foregoing that there is a need for a simple apparatus capable of imposing high rates of shear on oil solutions of polymers, thus enabling comparative tests to be carried out on commercially available materials, on production samples, and on research samples of new materials. Such an apparatus has been constructed and has been in use for some time in these laboratories.

#### THE APPARATUS

The apparatus was required to satisfy the conditions set out below.

In the first place it had to be of a robust construction, suitable for frequent and varied usage, without suffering significant wear, with its attendant change in test conditions. It was desirable that the components should be commercially available, thus ensuring rapid repair, replacement, or duplication. It was also necessary to carry out comparative tests on a relatively small quantity of material in a matter of a few hours.

After some consideration, and a few trial experiments, it was decided that an apparatus fulfilling the above requirements could be assembled from standard fuel injection equipment as supplied by Messrs. C.A.V., Ltd., with relatively few modifications. In principle, the solution, which replaced the gas oil normally used, was pumped at high pressure through a narrow orifice, high rates of shear being set up in the orifice itself. As the apparatus was to be used for comparative tests, no investigation of the type of flow in the orifice was carried out. However, it was possible to make a direct comparison of results obtained by Morris and Schnurmann under carefully controlled conditions, and those obtained in the present rig, using the same test fluid.

As the results were comparable in each case, and as Morris and Schnurmann have been able to calculate that their apparatus is capable of imposing a rate of shear of the order of  $10^7 \text{ sec}^{-1}$  it is considered that the present rig is capable of imposing a rate of shear of at least the same order.

As the materials to be tested were all of similar viscosity characteristics, no attempt was made to control the temperature of the apparatus during operation. It was found, however, that during operation the temperature of the solution, measured in the filter, rose rapidly to approximately  $38\text{--}40^\circ \text{C}$  and remained steady throughout the test. Thus the conditions set up in the apparatus were comparative in all cases.

At a later date several lubricating oil blends were investigated, for which purpose a small heating coil, controlled by an energy regulator, was wound round the outside of the filter, and by this means the temperature of the solution could be controlled within narrow limits over a wide range.

The apparatus (Fig. 1) consists of a fuel filter, pump and atomizer, working as in standard practice, the solu-

tion being returned to the filter after passage through the nozzle. The filter A, which also acts as reservoir, is connected via 3/16 in outside diameter thin-walled copper tube to the pump inlet chamber, a pet cock B being inserted as shown for withdrawal of samples. The filter element, as received, consisting of a wire cage covered with cloth, was found to cause a considerable "hold-up" of solution and was difficult to clean. Accordingly the cloth was replaced by a 200 mesh stainless steel gauze, which was found to be much more satisfactory in operation. The pump C, which is directly coupled to a one h.p. three-phase 440 r.p.m. electric motor, is a standard twin-cylinder model, from which one pump

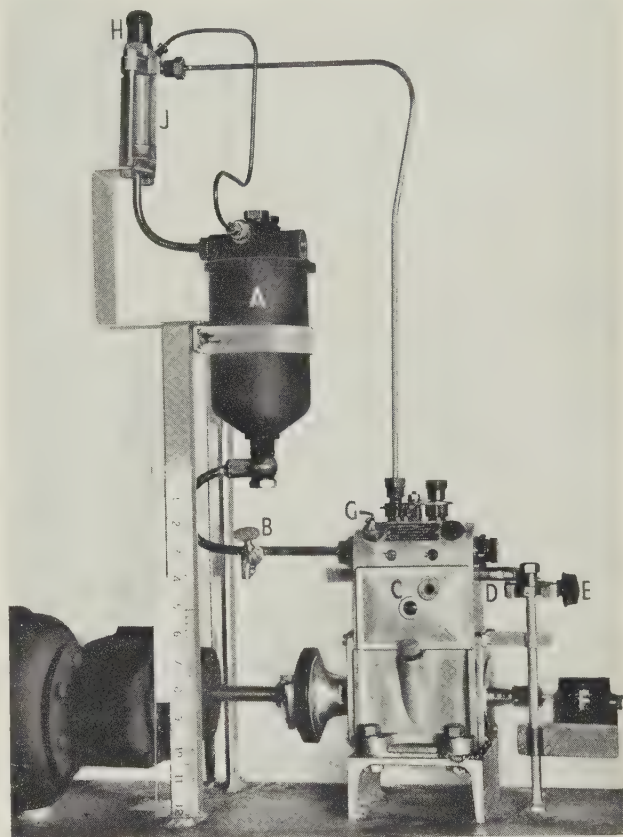


Fig. 1. General view of shear breakdown apparatus

element has been removed. The lower end of the cylinder is closed by means of a cork, which can be removed to facilitate flushing and cleaning of the inlet chamber and of the remaining pump element. The effective stroke, and thus the delivery of the pump, is controlled by rotation of the pump plunger in relation to the barrel, this being effected by means of the fuel rack, which in standard practice is connected to the throttle control and engine governor.

In the present case the fuel rack *D* is connected via a suitable link to a screw adjustment *E*, seen on the right of the pump (Fig. 1). Below this can be seen the revolution counter *F* which is directly affixed to the camshaft.

One of the pump "bleed screws" was drilled and tapped to take a miniature pet cock *G*, through which air, and incidentally solution, escape when the pump inlet chamber is purged of air. Solution is delivered, from the pump, at high pressure, through a length of 6 mm outside diameter  $\times$  2 mm inside diameter high-pressure tubing, to the injector and nozzle *H*, whence it is discharged into the de-aerator *J* and delivered finally to the filter. An exploded view of the injector and de-aerator

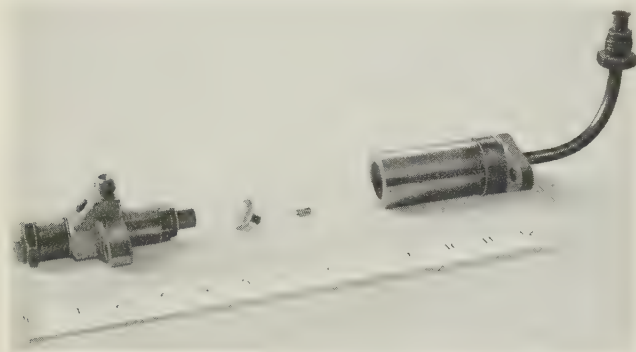


Fig. 2. Exploded view of injector and de-aerator

assembly is presented in Fig. 2, Fig. 3 being a cross-section of the de-aerator itself. As in standard practice, any solution leaking past the injector plunger is led from the "weep off," via 1/8 in outside diameter copper tube, to the filter. It is found that only a negligible amount of solution finds its way back to the filter by this route, thus by-passing the nozzle. A series of nozzles of different types was available, the "pintle" type being chosen because of its self-cleaning properties. In this type of nozzle the solution is ejected through an

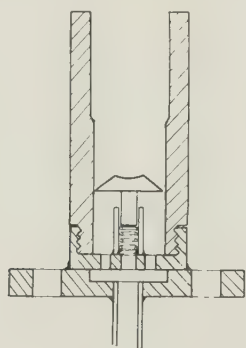


Fig. 3. Cross-section of de-aerator

annular orifice, the orifice being normally closed by a spring-loaded valve. When the pressure exerted by the solution on the valve is sufficiently high to compress the spring the valve opens, solution passes through the orifice, the pressure drops, and the valve reseats, closing the orifice once more. The pressure at which solution is permitted to pass through the orifice, the "breaking pressure," is controlled by varying the load on the spring controlling the valve. The moving parts of the pump

and injector, as received, were lapped to one micron and relied on the solution being pumped for their lubrication; thus it was essential to ensure that they never became dry and that the solution being pumped was free from abrasive matter at all times.

#### USE OF THE APPARATUS

The apparatus was calibrated using solutions of polymers in oil corresponding in viscosity to typical light and heavy hydraulic oils respectively.

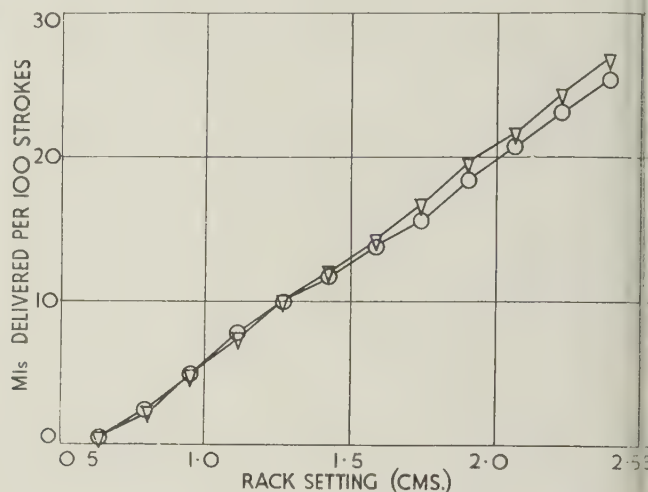


Fig. 4. Calibration of the apparatus

▽ High viscosity solution.  
○ Low viscosity solution.

The injector was cleaned by flushing with gas oil and set to the working pressure of 2500 lb/in<sup>2</sup> using a standard hand test set. The pump was cleaned and flushed with gas oil and the apparatus assembled, the de-aerator being arranged to deliver directly into a measuring cylinder. A suitable quantity of solution was transferred to the filter, the pump bleed cock opened, and solution collected during expulsion of air from the inlet chamber returned to the filter. The fuel rack was set to some suitable position and the apparatus started. It was permitted to run until approximately 20 ml of solution had collected in the measuring cylinder, thus ensuring replacement of gas oil by solution on the working parts of the pump and injector. The apparatus was then stopped, the solution in the measuring cylinder discarded and the revolution counter set to zero. The motor was restarted and allowed to run until a suitable volume of solution had collected in the measuring cylinder; the motor was then stopped, the volume of liquid collected and the revolution counter reading being noted. This procedure was repeated over the full range of rack settings for both types of solution. The results were plotted as ml of solution delivered per 100 pump strokes against rack setting. The graphs obtained are reproduced in Fig. 4. When shear resistance tests were carried out a known volume of solution was in circulation and



one time; thus reference to the calibration charts enabled the operator to determine how many pump revolutions were necessary to cycle the whole volume of solution once through the nozzle.

Tests were carried out in the following manner. A suitable quantity of solution (usually 250 ml) was pumped to the filter, the injector breaking pressure having been set to some predetermined value. Approximately 20 ml of solution were allowed to escape from the pump "bleed cock" during removal of air from the pump inlet chamber. This quantity of solution was returned to the filter. The rack was then set to the required position and the revolution counter reset. The motor was started and allowed to run until the whole volume of solution had made one complete cycle of the apparatus, thus ensuring thorough mixing of the solution and residual gas oil.

The motor was then stopped, a 100 ml sample of solution drawn off, the revolution counter reset, and the motor restarted. The motor was then allowed to run until the solution had performed the requisite number of cycles through the nozzle. At the end of the test a further 100 ml sample was taken and the viscosity of both samples determined in a standard Ostwald viscometer at some standard temperature. The duration of a test using 250 ml of solution for 50 cycles through the apparatus was of the order of two hours. The results are quoted as percentage viscosity decrease of the solution using the viscosity of the solution after one cycle through the apparatus as a base line.

In certain cases the polymer was recovered from the oil solution after test and its intrinsic viscosity determined in benzene at  $25 \pm 0.005^\circ \text{C}$ .

## RESULTS

Table 1. *The effect of high rates of shear on low viscosity solutions of polymers in gas oil*

Injector breaking pressure 2500 lb/in<sup>2</sup>

Polymer	Original viscosity of solution cs. at 54° C	Rack setting (cm)	No. of cycles	Percentage viscosity reduction at 54° C	Intrinsic viscosity of polymer in benzene at 25° C
A	10.45	1.75	1	0	0.3400
A	10.45	1.75	25	7.2 <sub>8</sub>	0.2937
A	10.45	1.75	25	6.9 <sub>7</sub>	—
A	10.45	1.75	50	10.3 <sub>4</sub>	0.2876
A	10.45	1.75	50	10.7 <sub>2</sub>	—
A	10.45	1.75	100	12.4 <sub>4</sub>	0.2703
A	10.45	1.75	100	12.7 <sub>0</sub>	—
B	11.25	1.75	1	0	0.2969
B	11.25	1.75	50	10.1 <sub>3</sub>	0.2203
B	11.25	1.75	50	9.2 <sub>2</sub>	—
C	10.25	1.75	1	0	0.2736
C	10.25	1.75	50	7.0 <sub>2</sub>	0.2100
C	10.25	1.75	50	6.5 <sub>4</sub>	—
D	10.02	1.75	1	0	0.9433
D	10.02	1.75	50	39.8 <sub>2</sub>	0.4278
D	10.02	1.75	50	39.9 <sub>0</sub>	—

A typical series of results obtained for solutions of various commercially available polymers (weight average molecular weight 80 000–800 000) in gas oil is reported in Tables 1 and 2.

Table 2. *The effect of high rates of shear on high viscosity solutions of polymers in gas oil*

Injector breaking pressure 2500 lb/in<sup>2</sup>

Polymer	Original viscosity of solution cs. at 99° C	Rack setting (cm)	No. of cycles	Percentage viscosity reduction at 99° C	Intrinsic viscosity of polymer in benzene at 25° C
E	10.98	1.75	1	0	0.3311
E	10.98	1.75	30	10.1 <sub>8</sub>	0.3021
E	10.98	1.75	30	10.3 <sub>5</sub>	—
E	10.98	1.75	50	13.0 <sub>4</sub>	—
E	10.98	1.75	50	13.1 <sub>0</sub>	—
E	10.98	1.75	100	15.2 <sub>7</sub>	—
E	10.98	1.75	100	15.5 <sub>0</sub>	—
E	10.98	1.75	150	16.0 <sub>6</sub>	0.2711
E	10.98	1.75	150	16.2 <sub>3</sub>	—
E	10.98	1.11	1	0	0.3311
E	10.98	1.11	25	7.9 <sub>7</sub>	—
E	10.98	1.11	25	7.9 <sub>3</sub>	—
E	10.98	1.11	50	11.9 <sub>1</sub>	—
E	10.98	1.11	50	11.6 <sub>5</sub>	—
F	10.00	1.75	1	0	0.3111
F	10.00	1.75	25	19.6 <sub>4</sub>	0.2660
F	10.00	1.75	25	19.0 <sub>3</sub>	—
F	10.00	1.75	50	23.6 <sub>5</sub>	—
F	10.00	1.75	50	24.2 <sub>7</sub>	—
F	10.00	1.75	111	28.7 <sub>2</sub>	—
F	10.00	1.75	111	29.3 <sub>4</sub>	—
G	9.37	1.75	1	0	0.4330
G	9.37	1.75	50	28.9 <sub>2</sub>	0.2821
G	9.37	1.75	50	29.5 <sub>9</sub>	—

A further series of experiments was carried out using a single hole nozzle, orifice diameter 0.9 mm, length 2.4 mm, in which the "breaking pressure" of the injector was varied. A few typical results are shown in Table 3.

The results quoted in Tables 1 and 2 are shown graphically in Figs. 5, 6 and 7. Reference to Figs. 5 and 6 shows that plots of percentage viscosity decrease as a function of the number of cycles through the apparatus are of similar shape to those obtained by Morris and Schnurmann,<sup>(2)</sup> thus confirming their results.

Table 3. *The effect of "breaking pressure" on the permanent viscosity loss of solutions of polymers in gas oil*

Rack setting 1.75 cm

Original viscosity of solution at 99° C = 10.54 cs

No. of cycles = 100

Polymer	Breaking pressure lb/in <sup>2</sup>	Percentage viscosity decrease at 99° C
E	2500	20.5 <sub>7</sub>
E	2500	21.0 <sub>1</sub>
E	1500	17.6 <sub>4</sub>
E	1500	17.3 <sub>8</sub>
E	1000	16.7 <sub>0</sub>
E	1000	16.9 <sub>2</sub>

It is of interest to note that the curves obtained in the present case are of the same general shape as those obtained by Millane,<sup>(4)</sup> and Staudinger *et al.*<sup>(5)</sup> utilizing a mill, and by Schmid and Rommel<sup>(6)</sup> and Mark<sup>(7)</sup> when investigating the effect of ultrasonic waves on polymer solutions.

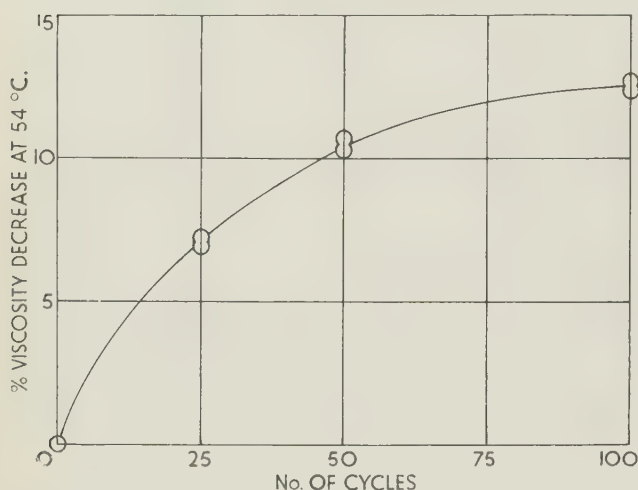


Fig. 5. Effect of shear stress on low viscosity solutions

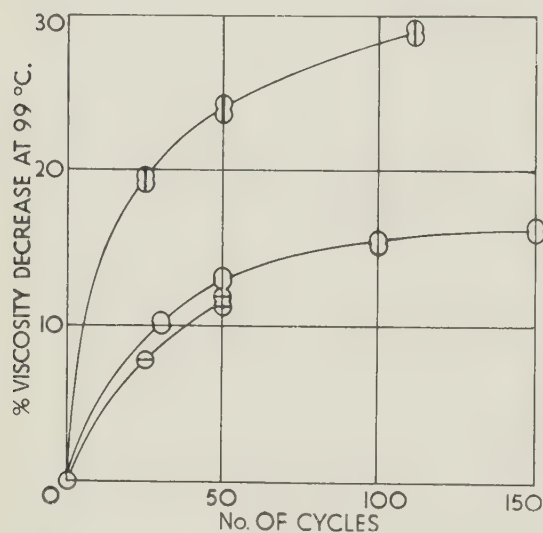


Fig. 6. Effect of shear stress on high viscosity solutions

- ⊖ Polymer F.
- Polymer E. Rack setting 1.75 cm.
- ⊖ Polymer E. Rack setting 1.11 cm.

A plot of intrinsic viscosity of the polymer as a function of the number of cycles through the apparatus (Fig. 7) gives a similar type of curve again, showing that, for a given rate of shear, only those molecules having a chain length greater than a certain critical value are ruptured, and that when all such molecules have been ruptured no further decrease in chain length of the polymer, and hence in the viscosity of the solution, will take place.

The results quoted in Table 3 show that, as was to be expected, a decrease in breaking pressure of the injector must lead to a decrease in the shearing stress and hence a smaller percentage reduction in the viscosity of the sample.

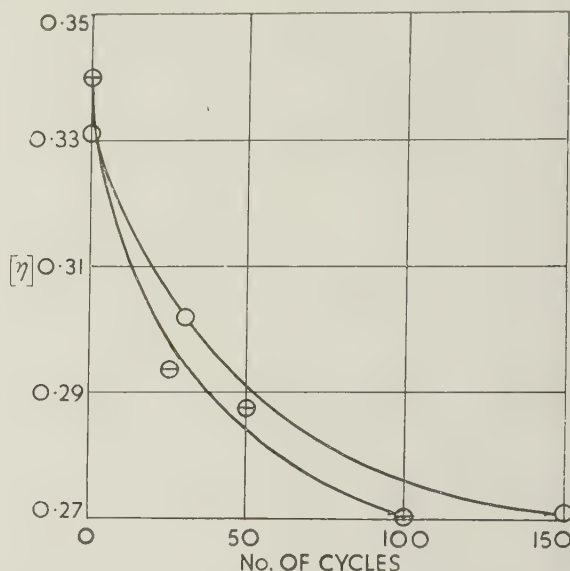


Fig. 7. Intrinsic viscosity of polymer solution as a function of number of cycles

- ⊖ Polymer A.
- Polymer E.

That the apparatus gives repeatable results is shown by the fact that several repeat experiments have been carried out recently after a lapse of more than twelve months, during which the apparatus was in constant use, and the results obtained were in all cases comparable with those obtained earlier. It should also be noted that no replacement of pump element or nozzle has been necessary.

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# The Rheology of Unseasoned Linoleum

## The Significance of Fillers in Calendering

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Kirkcaldy

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In addition to the formation of the sheet, the production of multi-coloured calendered linoleum of desired pattern is dependent on the plastic flow of the unseasoned material. By extruding the unseasoned linoleum mix in the apparatus described, its behaviour under calendering conditions and the effect exerted by the conventional fillers (cork, wood flour, and whiting) can be demonstrated with curves showing extrusion rates at different temperatures. A relationship is derived between the volumes of the different fillers required to give linoleum mixes of the same flow characteristics, and this relationship holds over a wide range of temperature and rate of extrusion. A few curves obtained with the apparatus for allied floorcoverings, e.g. asphalt tile and P.V.C. flooring are also given.

though built up on an empirical basis, the calendering of linoleum has been remarkably successful, and this has been achieved to a great extent by confining the composition of the mix and the mechanical variables within comparatively narrow limits, which long experience or trial and error have shown to give satisfactory results.

Modern mass production methods demand that more precise information be available on the properties of the unseasoned linoleum mass. Compounded from filled drying oil, rosin and fillers—both organic and inorganic—linoleum, prior to its formation into the familiar sheet form, is a plastic mass. The relative quantities and qualities of its ingredients determine its behaviour under the shearing stress of the calendering operation, and the thermoplastic nature of the linoleum renders this behaviour a function of temperature.

### PLASTOMETERS IN THE RUBBER INDUSTRY

Owing to the toughness of rubber and its compounds, rubber manufacturers have long been faced with the problem of assessing the rheological properties of the mixes they use and a variety of so-called plastometers employed throughout that industry. It was thought that some useful information on the linoleum compound might have been gained with one of these. Very briefly, rubber plastometers can be divided into three classes:—  
(i) Those in which a pellet of the material is compressed between parallel plates, as exemplified by the Williams plastometer. Owing to the difficulties of producing pellets of linoleum material that would not split under parallel plate pressure, it was decided that this type was not likely to give the required information for the linoleum calendering process.

(ii) The extrusion type. Originally suggested by Marzetti<sup>(1)</sup> working at comparatively low air pressure, and the modern type capable of working at many hundreds of lbs pressure, such as the Dillon<sup>(2)</sup> which is mainly used for tough extrusion mixes.

(iii) The shearing disk. The best-known instrument

of this type is the Mooney<sup>(3)</sup> plastometer, consisting of a cylindrical rotor running at 2 r.p.m. in a concentric stator chamber. Both the rotor and the stator friction surfaces are serrated. The temperature of the material under test may be controlled by means of steam or electrically-heated platens. The test material is filled into the stator chamber, the rotor started, and the torque measured at intervals over a pre-determined period, usually half an hour. The instrument bears some resemblance to the torsion viscometers used for the measurement of the viscosity of liquids.

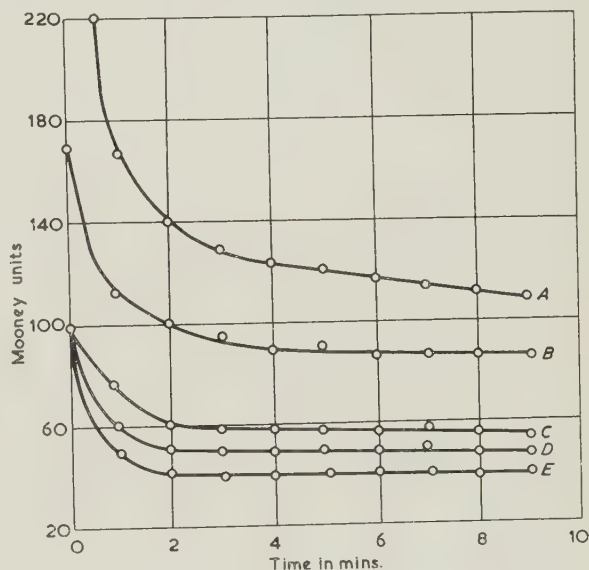


Fig. 1. Mooney plastometer curves for typical linoleum mixes

- A. A tough linoleum mix
- B. Same as "A" but containing more cement
- C, D, and E. All three are similar mixes with a constant volume of cement to filler, but the amount of whiting increases from "C" to "E" at the expense of the fibrous fillers

Typical curves of linoleum mixes, tested at 80°C in the Mooney plastometer, are shown in Fig. 1.

Fig. 1 shows that, after the initial mastication period, a fairly flat curve is obtained, suggestive of fluid viscosity. The position of the flat portions of the curves on the graph give an indication of the relative flow properties of the different mixes at the temperature of test. Linoleum mixes are, however, very sensitive to temperature change and thus, for the purposes of research, the Mooney instrument is not capable of giving the degree of temperature control required. Its possibilities for quick practical assessment in works control must not be overlooked.

#### THE EXTRUSION APPARATUS

The methods of investigation described are based on an apparatus used by A. G. Ward.<sup>(4)</sup> His results were of sufficient interest to warrant further investigation and this work is a corollary to his.

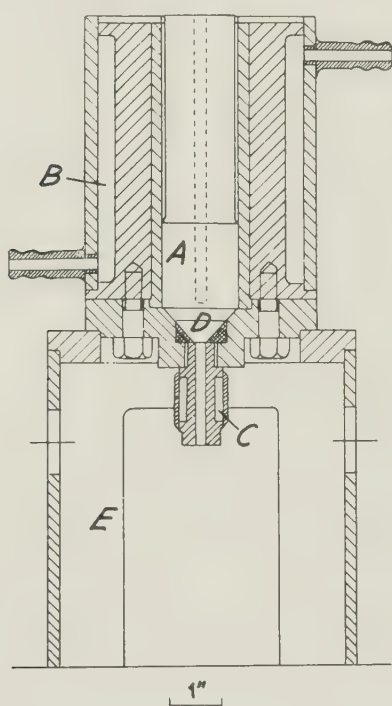


Fig. 2. Cut-away drawing of extrusion apparatus

- A. Piston with thermometer pocket
- B and C. Glycol circulation jackets. B and C are connected by flexible pipe
- D. Extrusion nozzle and tube
- E. Supporting stand for apparatus

The apparatus is essentially one in which a plunger forces the linoleum mass, under a controlled pressure, out of a cylinder, through a narrow tube, and is shown in Fig. 2. It consists of a steel cylinder,  $1\frac{1}{2}$  in. in diameter and 5 in. long, with a close-fitting piston. The length of the piston in actual contact with the cylinder is  $2\frac{1}{4}$  in. The wear surfaces of both the cylinder and the piston are case-hardened. To the bottom end of

the cylinder an extrusion nozzle is fitted, consisting of a hollow cone, leading into a tube  $\frac{1}{4}$  in. in diameter and 2 in. long. The sides of the conical lead-in from the cylinder to the tube are at an angle of  $45^\circ$  to the vertical. Both the cylinder and the nozzle are jacketed for accurate temperature control purposes. The apparatus is heated by circulating ethylene glycol with a centrifugal pump through the jackets at a pressure of 5 lb/in<sup>2</sup>. The ethylene glycol is heated electrically and, by thermostatic control, can be supplied at any desired temperature. Thermometer pockets are fitted to both the inlet and outlet pipes of the jackets and, in addition, a pocket is drilled in the piston itself reaching to within  $\frac{1}{8}$  in. of its bottom surface. During temperature standardization tests of the apparatus, a further thermometer was inserted through the nozzle and embedded in the test material. The temperature of the test material is controlled within  $0.2^\circ$  C.

The extrusion apparatus is provided with a stand, allowing of easy access to the material extruded, and it is placed between the platens of a Carver laboratory-type hydraulic press. The gauge, reading accurately up to 2 tons/in<sup>2</sup> indicates the pressure on the ram supporting the bottom platen, and this pressure is transmitted to the piston. The pressure ram is  $1\frac{1}{4}$  in. diameter and the control pressure on the material under test in the cylinder may be calculated from the gauge reading. A typical pressure calculation is shown.

Pressure required to lift ram and just to move the test material in the cylinder with nozzle removed = 95 lb/in<sup>2</sup>.

$\therefore$  Effective ram pressure is  $1\ 200 - 95 = 1\ 105$  lb/in<sup>2</sup>.

Actual pressure on extrusion piston is

$$\frac{(1\frac{1}{4})^2}{(1\frac{1}{2})^2} \times 1\ 105 = 768 \text{ lb/in}^2.$$

#### EXPERIMENTAL PROCEDURE

The material to be tested is mixed and sheeted to a gauge of 3 mm, in a laboratory 2-roll mill, by a standardized schedule corresponding to works practice. The sheeted material is cut into disks, using a  $1\frac{3}{8}$  in. diameter cork borer. A fixed volume of material, prepared from a pile of cut disks, is inserted in the cylinder of the apparatus. It is found that variations in the volume of material in the cylinder have little or no effect on the accuracy of the results obtained. The piston is inserted and sufficient pressure is applied to force some of the material in the cylinder through the nozzle. The apparatus is then left for a sufficient period for temperature equilibrium to be established.

When the correct temperature conditions are established, the apparatus is maintained at the desired constant pressure for a period sufficient to give measurable quantities of extruded material (which may be from 2 to 60 sec, depending on the properties of the material under test). Before each test, a 2 in. length is



extruded in order to remove any material which has been in contact with the air at the nozzle exit. The first test extrusion is discarded and further extrusions are made at three-minute intervals, the nozzle being cleaned each time, until six lengths of material are obtained.

The three-minute interval between each test extrusion allows time for temperature equilibrium to be re-established, should this have been disturbed by the movement of the plug in the cylinder. These extruded pieces are weighed and density determinations carried out, and the volume extruded in unit time is calculated for the conditions of test. When readings at various temperatures at a fixed pressure are required, the temperature control is adjusted after each set of extrusions, allowing sufficient time for the apparatus to reach temperature equilibrium, and the experiment is repeated.

The results may be plotted as temperature/extrusion curves. It is of interest to note that little difference has been noted in the density of the material extruded in the experiment proceeds, nor does this differ from the density of the plug removed from the cylinder at the end of the experiment. In this way, temperature/extrusion rate curves may for any given pressure be obtained for many types of thermoplastic mixes. Similarly, extrusion rate/pressure curves may be obtained for materials at any given temperature.

The experiments described refer almost entirely to extrusion at a fixed pressure at variable temperatures.

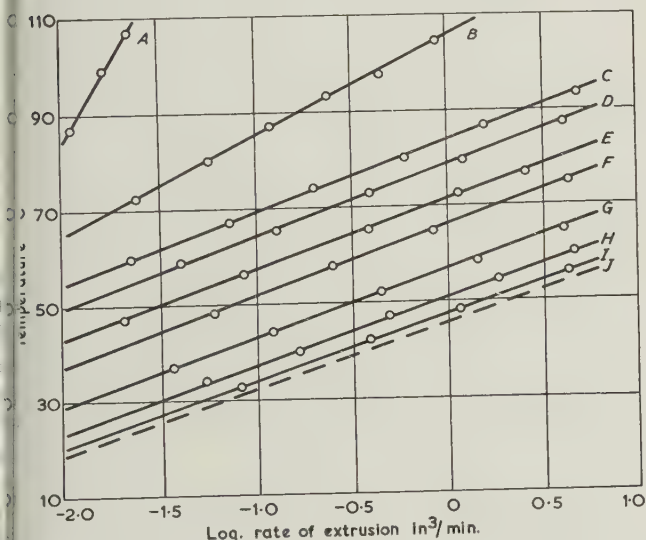


Fig. 3. Extrusion rate/temperature curves for linoleum cement/cork mixes

Gauge pressure 1 200 lb/in<sup>2</sup>

- |                         |                           |
|-------------------------|---------------------------|
| A. 30% binder by volume | F. 50% binder by volume   |
| B. 35% binder by volume | G. 61.6% binder by volume |
| C. 40% binder by volume | H. 70% binder by volume   |
| D. 43% binder by volume | I. 80% binder by volume   |
| E. 46% binder by volume | J. 100% binder by volume  |

Theoretical value derived from Fig. 4 and similar curves at other extrusion rates.

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## EXPERIMENTAL RESULTS

The three most widely used fillers in linoleum composition are cork, whiting and wood flour. Each of these materials has a particular effect on the calendering properties of a linoleum mix.

The apparatus described was used in an attempt to assess the relative effects of these fillers. The results are described. Linoleum cement (the linseed oil binder) from the same batch was used throughout the experiments. The sieve analyses of the fillers are given in Table 1, together with their densities, the latter figure being determined by the dead-space method.<sup>(4)</sup>

Table 1. Sieve Analyses—Cumulative Percentages

Retained on mesh	Cork	Wood flour	Whiting	Linoleum cement
42	8.1	Trace	—	—
60	36.3	9.6	—	—
80	55.7	38.8	—	—
115	68.1	76.2	—	—
170	78.7	97.1	1.9	—
Through 170	21.3	2.9	98.1	—
Absolute density	1.25	1.41	2.72	1.08

A series of mixes with the three fillers in turn was prepared, using various ratios by volume of binder and filler. Mixes containing less than 20% by volume of filler were difficult to extrude, owing to their sticky nature. The extrusion pressure was constant throughout the series of tests at a gauge reading of 1 200 lb/in<sup>2</sup>. Temperature extrusion rate figures were obtained for each mix and plotted as logarithm extrusion rate against temperature. The results for linoleum cement/cork mixes are shown in Fig. 3. Curves of a similar type were obtained for whiting and wood-flour mixes.

The straightness of the lines obtained is striking. Extrapolation gives figures for temperature outside the practicable capabilities of the instrument, e.g. where the extrusion rate is too small to be accurately measured.

Table 2. Relation between Extrusion Rate and Volume and Weight Factor

Extrusion rate (in <sup>3</sup> /min)	Fillers	Volume factor	Weight factor
0.1	Cork/whiting	1.16/1	0.53/1
	Cork/wood flour	1.29/1	1.14/1
	Whiting/wood flour	1.11/1	2.14/1
1	Cork/whiting	1.17/1	0.54/1
	Cork/wood flour	1.28/1	1.14/1
	Whiting/wood flour	1.105/1	2.13/1
5	Cork/whiting	1.20/1	0.55/1
	Cork/wood flour	1.33/1	1.18/1
	Whiting/wood flour	1.11/1	2.14/1

It will be noticed that, for any one filler, the curves are parallel for all ratios of filler to binder. There is, however, a tendency for the slope to become steeper at high filler concentrations. A possible explanation is

that the amount of binder is inadequate to wet the filler at those high concentrations, which, in any case, are beyond those used in manufacturing practice. Even experimentally they are difficult to compound.

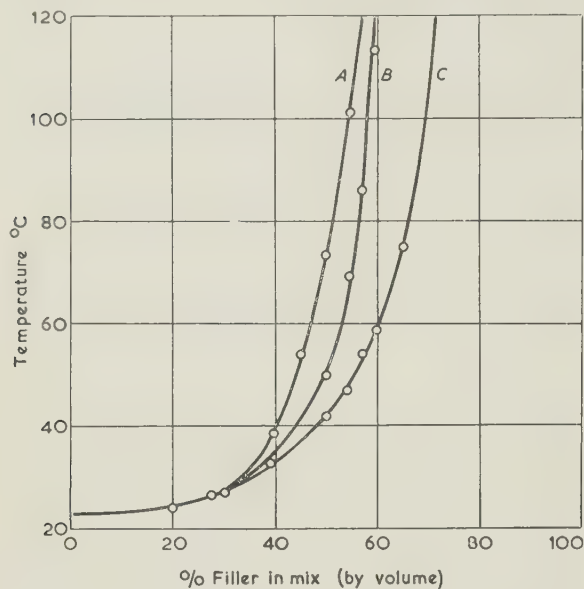


Fig. 4. Filler relationships with linoleum cement binder

Extrusion rate  $0.1 \text{ in}^3/\text{min}$   
Gauge pressure  $1\,200 \text{ lb}/\text{in}^2$

- A. Wood flour/linoleum cement mixes
- B. Whiting/linoleum cement mixes
- C. Cork/linoleum cement mixes

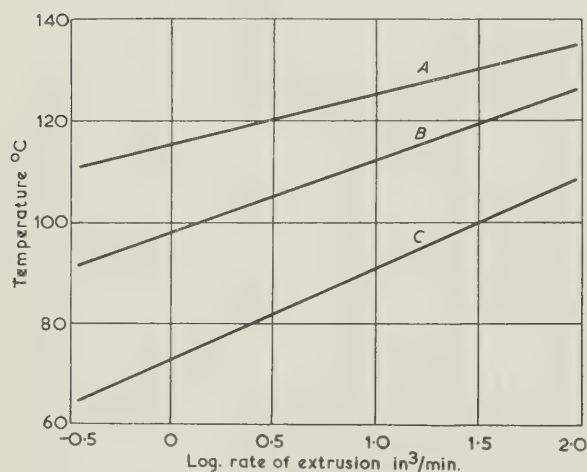


Fig. 5. Extrusion rate/temperature curves for hard linoleum, mastic tile and a typical P.V.C. flooring mix

Gauge pressure  $2\,000 \text{ lb}/\text{in}^2$

- A. Typical P.V.C. flooring mix
- B. Mastic tile mix
- C. Hard linoleum mix

From the temperature/log extrusion graphs, plots of temperature/filler content can be made for any selected extrusion rate. The rates of extrusion chosen were  $0.1$ ,  $1.0$  and  $5.0 \text{ in}^3/\text{min}$ . The curves for an extrusion rate of  $0.1 \text{ in}^3/\text{min}$  are shown in Fig. 4. Similar curves are obtainable with the other extrusion rates.

From these curves, by direct measurement, the relationships shown in Table 2 may be calculated. The figures quoted are the average figures taken at different temperatures over the range  $30^\circ/100^\circ \text{ C}$ . The relationship over the wide range of extrusion rates given is very close. The significance of the relationships obtained is that they enable one to replace, either wholly or in part, a filler in a linoleum mix by another and still retain the same rheological properties of the mass.

#### CONCLUSION

The results given demonstrate that, with the extrusion apparatus described, valuable data can be obtained on one of the most important properties of linoleum at the processing stage. In particular, the effect on flow properties exerted by the different conventional linoleum fillers is demonstrated. The accuracy attained is sufficient to permit a forecast of the behaviour of unseasoned linoleum mixes during manufacture. Alternatively, linoleum recipes from preferred ingredients can be immediately devised on theoretical grounds to suit plant conditions, without recourse to large-scale trial and error. By using a standard filler combination, it is possible to examine the properties of linoleum cements of varying compositions and physical state. There is also the obvious possibility of assessing and recording the properties of a mix which is known to give satisfactory performance on the plant.

No attempt has been made to deal with static friction or "stickiness" which also has its importance in linoleum sheet formation.

#### ACKNOWLEDGMENT

We are indebted to Dr. W. J. S. Naunton of the Imperial Chemical Industry Rubber Service Laboratory, Blackley, Manchester, for carrying out tests on the Mooney and Dillon plastometers.

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# Johnsen-Rahbek Effect with an Electronic Semi-Conductor

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The force of attraction which appears when a potential difference is applied between a piece of metal and a piece of semi-conductor, having accurately plane surfaces in contact with one another, has been investigated. Using an electronic semi-conductor the effect does not require the presence of moisture as Johnsen and Rahbek's experiment did. The force is very roughly proportional to the square of the applied potential.

Johnsen and Rahbek<sup>(1)</sup> investigated the attractive force which appears when a potential difference is applied between a piece of metal and a piece of lithographic stone, both having accurately plane surfaces placed in contact with one another. They found, among other things, that this force depends on the presence of a small amount of moisture in the lithographic stone and is presumably associated with the conduction induced by its moisture. Similar results have been obtained with slate, marble, flint, ivory, etc. Since the amount of moisture present is liable to vary with atmospheric conditions the effects are not very reliable and not suitable for practical application.

The author has made a similar investigation with an electronic semi-conductor, which does not rely on the presence of moisture for its conduction. Such experiments appear to have been carried out also by Weise.<sup>(2)</sup> Disks pressed from a powdered mixture of magnesium and titanium oxides, fired and reduced by heating in hydrogen to produce a suitable conductivity, were sprayed with metal on one face and soldered to metal blocks. The other face of each disk was then lapped using carborundum of various grades and finally polished, the polished surface being tested against an optical flat and interference-fringes. The disk was then secured to a base-plate with its polished surface uppermost.

A similarly lapped and polished brass block was laid, with its polished surface downwards, on to the semi-conductor disk and d.c. potentials applied between them. Attractive forces, of a similar order to those found by

Johnsen and Rahbek, were observed which disappeared when the potential was removed.

Details of the disks used are shown in the table. Column 2 indicates the degree of flatness of the disks in terms of the number of fringes observed against the standard flat. Column 3 gives the "average roughness" indicated by a Talysurf meter. Surface contour plots made by this machine on the disks indicated that small-scale irregularities of the order of  $10^{-4}$  cm were frequent. The bulk resistance of each disk was measured by dipping the lapped surface in a trough of mercury to form one electrode, the other being the block soldered to the metal-sprayed opposite face. The measured resistances are given in column 5 of the table.

The force required to lift the brass block from the semi-conductor was measured by means of a spring balance, correcting for the weight of the brass block, the average of six or twelve readings at each voltage being taken. A considerable scatter of results was found, part of the variation could be correlated with the manner of placing the disks together, but even when a standard procedure was followed fluctuation between successive observations still persisted. It was also observed that, if the pull was increased rapidly, somewhat higher readings were obtained than if it were increased slowly. This may have been due to "wringing" between the surfaces. These facts must be borne in mind when considering the average curves plotted in the figure, which should be regarded as having a rough, rather than precise, significance.

To establish whether the attractive force required the presence of moisture, as in Johnsen and Rahbek's

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## Details of Disks Used

Area of all semi-conductor disks was  $3.0 \text{ cm}^2$

Disk	Interference test	"Average roughness" (cm)	Condition	Resistance (MΩ)	$t$ calculated (cm)
A	3 fringes convex	$4.7 \times 10^{-5}$	Before drying	—	—
			After drying	0.20	$10 \text{ to } 16 \times 10^{-5}$
B	3 fringes concave	$5.9 \times 10^{-5}$	Before drying	—	—
			After drying	0.16	$7 \text{ to } 13 \times 10^{-5}$
C	4 fringes concave	$4.9 \times 10^{-5}$	Before drying	0.15	—
			After drying	0.20	$9 \text{ to } 11 \times 10^{-5}$
D	7 fringes concave	$2.5 \times 10^{-5}$	Before drying	1.0	—
			After drying	2.5	$23 \text{ to } 26 \times 10^{-5}$
E	4 fringes concave	$4.6 \times 10^{-5}$	Before drying	2.0	—
			After drying	2.5	$11 \text{ to } 13 \times 10^{-5}$
Brass	3 fringes convex	—	—	—	—

experiments, four of the disks were heated to about 80°C, then kept in a dessicator for several days and tested immediately after removal from it. This treatment did not diminish the force of attraction and with two of the disks the forces were considerably higher afterwards.

The simplest explanation of the attractive force is that it is the normal electrostatic attraction between the surfaces. Actual electrical contact will only occur at a very few regions of small area, elsewhere there will be a film of air of average thickness about  $10^{-4}$  cm between the surfaces. When a voltage is applied across the system the current must pass through the small contact regions which form the major part of the resistance of the circuit, so that most of the potential drop occurs very near these regions, and the potential drop elsewhere is negligible. The two surfaces separated by only about  $10^{-4}$  cm, therefore, have a difference of potential between them nearly equal to the externally applied potential, except very near the contact regions, and attract one another according to the well-known law of electrostatics. Making the simplifying assumption that the separation and the potential difference are uniform over the whole surface, we have:

$$t = \left(\frac{A}{8\pi}\right)^{\frac{1}{2}} \frac{V}{300\sqrt{Wg}} = 3.7 \times 10^{-5} \cdot \frac{V}{\sqrt{W}}$$

where  $A$  = area in  $\text{cm}^2$  ( $3.0 \text{ cm}^2$ );

$W$  = force in grams weight;

$V$  = potential in volts;

$t$  = width of gap in cm;

$g = 981 \text{ cm sec}^{-2}$ , acceleration due to gravity.

If the law of electrostatic attraction were accurately obeyed the force/voltage curve shown in the figure would be straight lines with a slope of two. Since they are not, a different value of  $t$  will be found for each readings, presumably because the separation varies with the applied voltage, or because the potential difference across the gap is not proportional to the voltage. Column 6 of the table shows the range of values of  $t$  found by applying the equation given above to the results for each disk. Disk  $B$  which, being concave to about the same extent as the brass disk is convex, might be expected to fit it best, does in fact give the lowest values of  $t$ , of the same order as the roughness.

Disks  $A$  and  $D$ , on the other hand, are expected to misfit by two or three half-wavelengths and  $t$  turns out to be about two wavelengths for the former and four for the latter.

It will be seen from the figure that the observed force is far from accurately proportional to the square of the voltage. The following effects might account for deviations:

(1) Change of resistance of the contact region with change of the voltage (and current) due to local heating, to increase of area by compression, or, less probably, to barrier layer effects.

(2) Distortion or compression of the surfaces. The force is measured when the surfaces are about to separate, so that the net pressure on them should be zero, but the combination of electrostatic pull and the opposing pull of the spring balance may alter their configuration, and so alter the effective separation.

(3) Crushing or plastic deformation. It seems possible that the surfaces may be held apart on a number of tiny pinnacles which may penetrate the metal or be partially crushed during the application of the electric field.

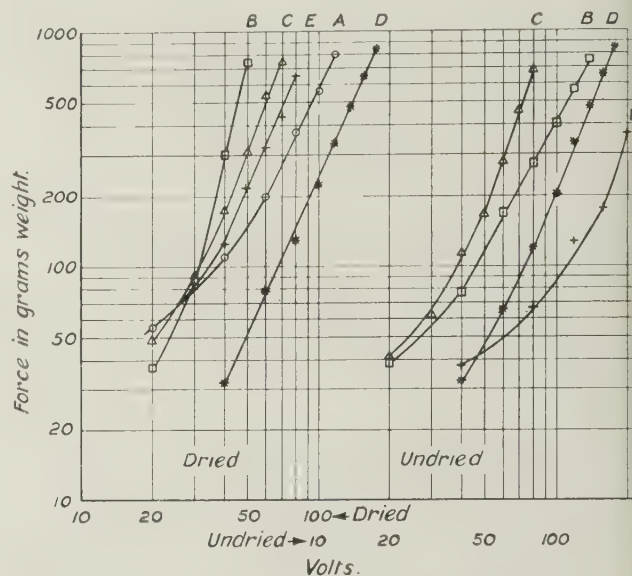


Fig. 1. Relation between force and voltage

The first of these would tend to lessen the rate of increase of force with voltage, the other two would tend to increase it. Most of the curves shown suggest that the former effect is outweighed by the latter at the higher voltages.

The considerably increased attractive force which appeared with two of the disks after heating and drying in a dessicator is not easy to explain. It is possible that the heating to 80°C may have altered the surface configuration favourably, or on the other hand it may have removed a layer of moisture which was increasing the conductivity near the surface and affecting the potential distribution unfavourably.

The currents flowing between brass and semi-conductor were measured for various voltages. For an applied voltage of about 40 V the current varied from about  $0.2 \mu\text{A}$  to about  $10 \mu\text{A}$  for the various disks depending upon the bulk resistance of the disk and the contact resistance at the junction. The current increased rapidly with the applied voltage, indicating a rapid decrease of contact resistance which may be due to local heating of the contact regions, to increase of contact area by compression, or to barrier effects. From other observations it appears unlikely that barrier effects are appreciable at these voltages. Those disks displaying



largest attractive force also showed the most rapid increase of current with voltage, suggesting that compression and enlarging of the contact areas may play an important part. The current at the lower voltages were consistent with contact regions being of the order of 0.1 to 0.1 cm diameter.

Although the explanations are far from complete, there is no evidence which clearly conflicts with the view that the force is a normal electrostatic one; while the high proportionality between the force and the square of the potential, and the reasonable values deduced for the separation between the surfaces, support this view.

## ACKNOWLEDGMENTS

The author records his thanks to Sir Arthur Fleming, Director of Research and Education, and to Mr. B. G. Churcher, Manager of the Research Department, for permission to publish this paper.

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## Correspondence

## The Diffusion of Moisture through Flax Seed

The problem outlined by Samuels and Tait (*Brit. J. Appl. Phys.*, p. 125, May 1950) is amenable to a little further analysis. Previous experience, confirmed by the authors, suggests that liquid flow of water from seed to seed is negligibly small, and the main transport of water through mass of seeds must be by diffusion of water vapour. The above equation is the same for both absorption and desorption:—

$$\frac{dm}{dt} = -D \frac{d\rho_v}{dx} \text{ gm/cm}^2/\text{sec.}$$

where  $m$  is the mass of water crossing 1 cm<sup>2</sup> per sec.,  $D$  is coefficient of diffusion of water vapour through the porous mass, and  $\rho_v$  is the concentration of vapour in gm water per c.c. of air in the pore space. (Note that these symbols are not the same as those of Samuels and Tait.) The value of  $D$  is less than that in free air ( $D_0$ , say where  $D_0 \doteq 0.25$  cm<sup>2</sup>/sec at ordinary temperatures), and if the flax seeds were spherical the ratio  $D/D_0$  could be derived from a curve published some years ago.\* As the seeds are flat, I have used the technique and equipment described in my paper to estimate  $D/D_0$ , and find it to be  $D/D_0 = 0.155$ . Hence  $D = 0.04$  cm<sup>2</sup>/sec.

From their Fig. 1, for the range of humidity and moisture content in the adsorption experiments

$$\rho_\omega/\rho = a + bh$$

where  $\rho_\omega$  is the density of water in gm per c.c. of bulk seed,  $\rho$  is the density of seed in bulk (measured on my samples as 0.70 gm/c.c.),  $a$  and  $b$  are constants, and  $h$  is the relative humidity (as a fraction never greater than unity). Obviously,  $\rho_v = \alpha h$ , where  $\alpha = 1.73 \times 10^{-5}$  for water vapour at 20° C.

Hence we may write

$$\rho_v = \alpha h = \frac{\alpha}{b} \left( \frac{\rho_\omega}{\rho} - a \right)$$

and the diffusion equation becomes

$$\begin{aligned} \frac{dm}{dt} &= -D \frac{d}{dx} \left[ \frac{\alpha}{b} \left( \frac{\rho_\omega}{\rho} - a \right) \right] \\ &= -\frac{D\alpha}{b\rho} \cdot \frac{d\rho_\omega}{dx} \end{aligned}$$

This is essentially equation 4 of Samuels and Tait, with their  $D_a$  replaced by  $D\alpha/b\rho$ . All values are known:  $D = 0.04$  cm<sup>2</sup>/sec,  $\alpha = 1.73 \times 10^{-5}$ ,  $b = 0.12$  (from Fig. 1), and  $\rho = 0.70$  gm/c.c.

$$\begin{aligned} \therefore D_a &= 8.3 \times 10^{-6} \text{ cm}^2/\text{sec.} \\ &= 0.72 \text{ cm}^2/\text{day.} \end{aligned}$$

Samuels and Tait find  $D_a = 0.74 \pm 0.16$  cm<sup>2</sup>/day.

A similar treatment for a non-linear moisture characteristic can be based upon the assumption of a second degree relation between  $\rho_\omega$  and  $h$  over the relevant range: say,

$$\rho_\omega/\rho = a + bh + ch^2$$

$$\begin{aligned} \text{i.e. } \frac{1}{\rho} \frac{d\rho_\omega}{dx} &= \frac{b}{\alpha} \cdot \frac{d\rho_v}{dx} + \frac{2c}{\alpha^2} \cdot \rho_v \cdot \frac{d\rho_v}{dx} \\ &= \frac{1}{\alpha} \cdot \frac{d\rho_v}{dx} (b + 2ch) \end{aligned}$$

and hence

$$D_d = D\alpha/\rho(b + 2ch)$$

where  $b + 2ch$  is the slope of the moisture characteristic at  $h$ .

(There is no need to separate cases as above. A general treatment is easy and shows that the diffusion coefficient as defined by Samuels and Tait is always inversely proportional to the slope of the moisture content/humidity curve. Although easy, the general treatment is not so revealing.)

Applying this result to desorption, the value of  $D_d$  decreases with increasing humidity. Calculating values for the extremes quoted by Samuels and Tait, they are: 0.157 and 0.430 cm<sup>2</sup>/day; Samuels and Tait find 0.148 and 0.470 cm<sup>2</sup>/day for 87% and 70% relative humidity respectively. The data plotted on their Fig. 5 should, therefore, lie between two straight lines, both passing through the origin, and having slopes in a ratio of about 1 to 3. That the data do in fact fall very nearly on a third straight line, almost coincident with the steeper of these two but not passing through the origin, is somewhat fortuitous, and is probably a result of a regular sequence in their handling of their data.

H. L. PENMAN.

Rothamsted Experimental Station, Harpenden, Herts.

\* PENMAN, H. L. *J. Agric. Sci.*, 1940, **30**, p. 437.

## NOTES AND NEWS

## New Books

**Symposium on Particle Size Analysis.** Edited by H. W. THORP. (London: Institution of Chemical Engineers.) Pp. 145. Price 42s. net.

The properties of fine particles excited the interest of the great classical physicists and they solved some of the basic problems, but the subject has since assumed such importance in industry that a considerable volume of research, both pure and applied, has been devoted to it in recent years. The Institution of Chemical Engineers, the Society of Chemical Industry, and Dr. H. Heywood, who organized the symposium and read a noteworthy paper, are to be congratulated on having collected together a series of papers of high merit from specialists in this field of work. Not only do the papers cover critical reviews of previous work, but they also contain much original matter.

The symposium was divided into two parts. The first dealt with methods of measurement and includes papers on the scope of particle size analysis and standardization, mono-dispersed systems, the theory of sedimentation of particles, determination of surface area by adsorption, heat of wetting, permeability and light extinction methods, and the electron microscopy of fine particles. In the second part, some industrial examples are considered and there are papers on particle size analysis in pigments, soils, and bitumen emulsions, measurements in the radio industry, and a new apparatus for use in the sub-sieve range. Although three years have elapsed since the symposium was held, this compilation has not lost significantly in usefulness, but opportunity might well have been taken to add later information to some of the papers even at the expense of pruning the discussion, valuable as this may be. For instance, a summary of recent publications on the application of the Mie theory to the scattering of light by small particles would have helped the reader to a better appreciation of the errors in light extinction methods.

The list of contents omits the communication on mono-dispersed systems and does not include the discussion, although references are given in small print at the end of each paper to the pages where the relevant remarks can be found. There is no index, misprints are too frequent, and the binding is poor. These blemishes are regrettable because the book will come into widespread use in research and industrial laboratories as representing the most comprehensive contribution to particle size analysis yet made in this, or indeed in any, country.

H. L. GREEN

**Report on the Principles of Rheological Nomenclature.**

By J. M. BURGERS and G. W. SCOTT BLAIR.  
(Amsterdam: North-Holland Publishing Co.)  
Pp. v + 72. Price f. 2.50.

It is impossible in few words adequately to review this report. It represents a courageous and thorough attempt to present a consistent system of rheological nomenclature and within its acknowledged limitations it succeeds. Having defined the scope of the report, the authors devote a chapter to the elements of stress and strain analysis. The classification following is based on the results of various experiments performed on the material. The magnitude of the permanent

deformation is considered and a "flow-diagram" (permanent shear against time) constructed. From these flow-curves a distinction may be drawn between elastic, plastic and fluid behaviour, and in cases of appreciable permanent deformation various types of flow (limited, uniform, decelerating, accelerating) may be considered. With uniform flow a further classification is possible into Newtonian, Bingham shear-rate thickening and thinning liquids. Chapter IV deals with recovery of shape, retardation effects, various types of elasticity, stimulated recovery and creep. The following chapter discusses work hardening and various types of thixotropic behaviour. Finally the terminology developed is compared with the nomenclature of other authors.

Rheological nomenclature is at present in chaotic state. As an example of stupid terminology the word "dilatant" as applied to systems showing increase of apparent viscosity with rate of shear may be cited: such systems often show no change in overall volume on stirring. Nomenclature is, however, a dull subject and it is tempting for the individual author to adopt the methods of Humpty Dumpty in his papers. In the interests of clarity and international co-operation, this temptation should be resisted. The present report marks an important step forward in the direction of satisfactory terminology and deserves close study.

It should be pointed out that the published report is now issued with a duplicated insert which forms no part of the report and has not met with general approval.

V. G. W. HARRISON

**Acoustic Measurements.** By L. L. BERANEK, S.D., D.Sc. (New York: John Wiley; London: Chapman and Hall.) Pp. 914. Price 56s. net.

Dr. Beranek is Associate Professor of Communications Engineering and Technical Director of the Acoustics Laboratory, Massachusetts Institute of Technology, and the book has been prepared under the auspices of, and through funds supplied by, the Office of Naval Research, Navy Department, Washington, D.C. In the preface the author states that the book is intended primarily as a reference for graduate students and workers in the field of acoustics. The subject is covered in such a way that "the book will be an aid to five main groups of research workers: the acoustical physicist making fundamental laboratory measurements, the communications engineer measuring and evaluating the performance of audio communication systems, the psychologist performing measurements of the human hearing mechanism, the otologist studying hearing defects, and finally, the industrialist applying acoustic measuring techniques in manufacturing processes."

This comprehensive volume deals with the subject in series of twenty chapters. It is impossible to refer to these individually in a brief review, but their main contents may be of interest. The opening chapter gives a brief historical review of the development of the subject from the earliest times, followed by an alphabetical list of definitions of acoustical terms now in general use. This is followed by discussion of the acoustical characteristics of the medium, notably the atmosphere, in which sound is propagated. The important aspects of attenuation, scattering and non-linear



stortion in wave propagation are discussed. The diffraction of sound waves around various obstacles (spheres, cylinders and the human body) and around sound sources in finite baffles, are dealt with in relation to the wavelength of the sound employed. A chapter on the primary techniques for the measurement of sound pressure and sound velocity and for the absolute calibration of microphones includes discussions on the "reciprocity" technique of calibrating microphones, the theory and use of the Rayleigh disc, concluding with a description of some primary sources of sound (the thermophone, pistonphone, and the electrostatic transducer). An interesting chapter follows on microphones and ears, in which Fletcher's curves of equal loudness contours and various types of microphone (carbon, condenser, moving coil, piezo-electric, directional, etc.) are discussed. In dealing with the important subject of frequency measurement, the various types of frequency standard are discussed, viz. (1) primary (e.g. crystal clocks); (2) laboratory (pendulum clocks, precision forks); (3) ordinary (audio oscillators, electronic meters, Wien bridge, etc.); (4) frequency comparison methods. In the following chapter the measurement of acoustic impedance (including motional electrical impedance) is discussed. Later chapters deal with the following subjects: the audiometer; sound sources for test purposes; characteristics of random noise; indicating and integrating instruments for the measurement of complex waves; analysis of sound waves; basic tests for communication systems—the fitting of microphones, amplifiers and loudspeakers; tests for laboratory and studio microphones; tests for loudspeakers; testing of communication system components; articulation test methods; measurement of the acoustic properties of rooms, studios and auditoriums, measurement of acoustical materials; the sound level meter.

There is no other book on acoustic measurements available which covers so wide a field. The author evidently intends the book to make a very wide appeal, but the wisdom of doing this is a little doubtful. In view of the large size and price of the book it might have been better perhaps to publish the contents in either two or three separate volumes, each with its particular appeal to the groups of research workers mentioned in the preface. As it stands, sections which would appeal to a physicist or an acoustics engineer would not appeal to the psychologist or otologist, and conversely. As a physicist, the reviewer feels that much of the text could have been omitted or greatly condensed without serious loss, but no doubt the communications engineer and the psychologist would be most interested in precisely such omissions. There is no intention of criticizing in this way the contents of the book, which contains a mine of useful information, but merely to indicate that it contains a rather indigestible mixture for specialists in any one group mentioned by the author in the preface.

It is a little disappointing to find in a book sponsored by U.S. Office of Naval Research so little reference to measurement of underwater sound, but perhaps this is explainable by the secret nature of much of the underwater acoustic research carried out either in America or any other country. Nevertheless a good deal of information on such research has been published from time to time. Perhaps it is a little pedantic to criticize minor items in such a valuable treatise, but, to mention only one or two, I doubt whether most physicists would agree with Dr. Beranek's definitions of "nodes" (p. 29) as "points, lines, or surfaces (of pressure, velocity, or displacement) of a stationary wave system which

have zero amplitude," and "antinodes" (p. 16) as points, etc., "which have maximum amplitude of pressure or velocity." Surely a node in a stationary wave system is a point of minimum velocity and maximum pressure amplitude, these quantities being respectively maximum and minimum at the antinodes. Again, on p. 19, we find the definition that the "*Diaphragm* of an electroacoustic transducer is that portion which is actuated by sound pressures or from which sound is radiated." Whilst admitting the frequent use, or misuse, of the expression "piston diaphragm" in the case of a piezo-electric piston source or receiver, it requires a stretch of imagination to speak of the sound-emitting or receiving edge of a pile of nickel stampings in a magnetostriction oscillator as a diaphragm. The sub-heading 2.7 "*Diffraction and scattering*" on p. 75, a section dealing with temperature gradient effects in the atmosphere, should obviously read "*Refraction and scattering*."

The index, a very important part of any reference book, is frankly disappointing; references to important items which are mentioned in the text are missing or incomplete in the index. To quote one example only, "impedance—motional," the index mentions p. 25, where is found a definition only, whereas a useful account of methods for its measurement is given on pp. 351–61. Perhaps one should not criticize an American publication on the grounds of its lack of references to European publications, but the reader certainly gets the impression on reading the book that European literature on acoustics is not readily available to the author.

In spite of these minor criticisms, the book forms a very valuable reference, particularly to United States work in the field of acoustical measurements, and will be most appreciated by those engaged in the technological side of acoustics (acoustical and communication engineers). It is a most impressive volume showing how acoustics has developed into electronic engineering during recent years; on reading the book it is not easy to decide whether acoustics is a branch of electronics or vice versa!

The book is written by an author with wide practical experience in the field of acoustic measurements and bears the impress of close acquaintance with much of the work he describes. The volume is well reproduced on good paper and is profusely illustrated.

A. B. WOOD

**The Scientist's Ready Reckoner.** By W. ROMAN, Ph.D., A.F.Inst.Pet. (Netherlands: Dr. W. Junk, The Hague.) Pp. 144. Price 15s. net.

This type of publication is a form of numerical yarn-swapping. Most researchers have a personal collection of tables or data which it has been useful to compute or collate for frequent reference in a particular laboratory, and this handbook, by the chief analyst of Petrocarbon, Manchester, contains an extensive collection of information of this kind. The tables, mainly to four figures, include values (with their common logarithms) of atomic weights, multiples of atomic weights, weights of radicals and molecules, gravimetric factors and titrimetric equivalents. Specific gravities are given and there are also temperature conversion tables and common logarithm tables.

The book is probably of little personal use to the professional physicist, but may be of value to senior technicians and assistants in chemical laboratories. Although the standard of production is good, many of the tables would have been greatly improved by better arrangement and layout.

N. CLARKE

## Notes and Comments

## Elections to The Institute of Physics

The following elections have been made by the Board of The Institute of Physics:

*Fellows:* J. R. Atkinson, G. E. Bacon, H. Barrell, W. J. R. Calvert, D. W. Gillings, A. Hargreaves, K. R. Makinson, T. C. Marwick, M. Milbourne, G. Morris, T. L. Richards, J. B. Rudd, M. K. Sen, L. A. A. Thomas, H. Tompa, H. W. Thompson, C. G. Wynne.

*Associates:* W. H. Alexander, I. P. Bell, G. W. Bloomfield, W. R. Blunden, K. W. Bridger, R. Brooks, L. Brown, A. G. Cannon, J. Critchlow, A. W. Cronshaw, D. E. Davies, P. Denton, J. F. Dillon, G. W. Eastwood, D. Gifford, A. M. Goddard, R. H. Hall, E. L. Hanson, F. H. Hibberd, R. J. Hodgkinson, W. E. Hurley, D. Jolly, C. E. Jones, E. B. Jones, E. W. Jones, R. Joseph, H. D. Keith, A. J. Kent, J. B. Le Poole, A. G. Loudon, K. W. Lyon, C. R. Major, E. D. Malcolm, J. M. Messenger, A. C. Moore, J. Moore, D. A. Perrott, W. A. D. Randall, E. Riley, P. W. Roberts, B. N. Sen, H. S. Sowery, A. G. Thompson, N. S. Thumpston, J. G. Walford, W. F. Ware, E. J. W. Whittaker, G. J. Williams-Leir, D. J. Williams, J. W. Winstanley, B. A. Worswick.

Sixty-eight Graduates, five Subscribers and twenty-seven Students were also elected.

**X-ray Analysis Equipment.** The Equipment Sub-Committee of the X-ray Analysis Group of The Institute of Physics makes the following announcements regarding the work of its Panels:

(1) A new Graphical Methods Panel has been formed to deal with the supply of the different types of charts and nets used in X-ray analysis. The Secretary is Mr. A. E. De Barr, Research Laboratories, Elliott Brothers (London), Ltd., Elstree Way, Borehamwood, Herts.

(2) The Single Crystal Panel has been reconstituted and observations on the work of this Panel should now be addressed to the Secretary, Dr. W. Cochran, Crystallographic Laboratory, Cambridge.

(3) The Powder Camera Panel has been reconstituted and renamed the Powder Camera and Geiger Counter Spectrometer Panel. Additional members have been enrolled to provide a broad basis for consideration of Geiger counter work in X-ray crystallography. Suggestions are welcomed and should be sent to the Panel Secretary, Mr. H. S. Peiser, Messrs. Hadfields, Ltd., East Hecla Works, Sheffield, 9.

**Conference on Automatic Control, 1951.** A Conference on Automatic Control, sponsored by the Department of Scientific and Industrial Research, will be held from 16 July to 21 July, 1951. It will embrace the following main topics:

(1) Servo Control Systems: General Theory and Problems of Synthesis. (2) Servo Control Systems: Non-linearities. (3) Process Control: Inter-relation with Servo Control. (4) The Human Operator.

The Organizing Committee is under the chairmanship of Prof. A. Tustin, of the Department of Electrical Engineering, Birmingham University, who invites contributions. Potential contributors should submit a title and brief synopsis to the Secretary of the Committee, Mr. R. G. Silversides, at D.S.I.R. Headquarters, Charles House, 5-11, Regent Street, London, S.W.1, by 31 August, 1950. Papers selected for presentation will be required in their final form not later than 1 January, 1951. Only in exceptional circumstances will those in excess of 5 000 words be accepted.

**Instrument Industry's Exhibition.** The Scientific Instrument Manufacturers' Association of Great Britain, Ltd. (S.I.M.A.) announce that the first British Instrument Industry's Exhibition to be held will take place in the National Hall, Olympia, London, from 4-14 July, 1951. This important exhibition which deserves the warmest support from the scientific instrument and allied industries, will, we understand, occupy some 50 000 sq. ft. The exhibition is also being supported by the British Industrial Measuring and Control Apparatus Manufacturers' Association, the British Electrical and Allied Manufacturers' Association (Instruments Section), the British Lampblowing and Scientific Glassware Manufacturers' Association and the Drawing Office Material Manufacturers' and Dealers' Association, Ltd. It is also hoped that a section of the exhibition will be occupied by users of the instrument exhibited.

## Journal of Scientific Instruments

## Contents of the August issue

## ORIGINAL CONTRIBUTIONS

- A Continuous-Shear Rheometer for measuring Total Stress in Rubber-like Materials. By W. F. O. Pollett and A. H. Cross.
- An Accelerometer for measuring Ship Hull Vibrations. By A. G. Boggis.
- A Recording Double Beam Infra-red Spectrophotometer. By I. A. Brownlie.
- A Transportable Radiation Monitor. By K. Kandiah and M. O. Deighton.
- A Liquid Level Gauge. By E. W. R. Little.
- The Chemical Micro-Analysis of Binary Gas Mixtures. By F. C. Tompkins and D. M. Young.
- A Schmidt Mechanism for Approximate Solution of the Equation of Linear Flow of Heat in a Medium whose Thermal Properties depend on the Temperature. By J. C. Jaeger.

## LABORATORY AND WORKSHOP NOTES

- A Multiple High-Vacuum Valve Unit. By R. I. Garrod and R. A. Coyle.
- A Capacitance Variation Microinch Comparator. By G. W. Fynn.
- Laboratory Bench Fittings. By M. J. Moore.
- Electrically Operated Air-Inlet Valve. By A. J. Croft.
- The Preparation of Thin Layers of Uranium Oxide. By G. B. Cook and F. Hudswell.
- Insulated Power Lead-in for Vacuum Systems. By M. Moore.
- Direction of the Plane of Polarization of a Nicol Prism. By E. J. Burge.

## Notes and News

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**EDITORIAL MATTER.** Communications concerning editorial matter should be addressed to the Editor, The Institute of Physics, 47 Belgrave Square, London, S.W.1. (Telephone: Sloane 9806.) Prospective authors are invited to prepare their scripts in accordance with the *Notes on the Preparation of Contributions* which will be sent gratis on request.

**ADVERTISEMENTS.** Communications concerning advertisements should be addressed to the agents, Messrs. Walter Judd Ltd., 47 Gresham Street, London, E.C.2. (Telephone: Monarch 7644.)

**SUBSCRIPTION RATES.** A new volume commences each January, and annual subscriptions will only be accepted for complete volumes of the monthly parts (January-December). The charge is £3 per volume (\$8.50 U.S.A.), including index (post paid), payable in advance. Single parts, so far as available, may be purchased at 6s. each (85c. U.S.A.), post paid, cash with order. Orders should be sent to The Institute of Physics, 47 Belgrave Square, London, S.W.1, or to any Bookseller.



## SPECIAL REPORT

# Summarized Proceedings of Conference on "X-Rays in Industry"— Melbourne, Australia, November, 1949

A Conference on "X-Rays in Industry," organized by the Australian Branch of The Institute of Physics, was held on 9–11 November, 1949, in the University of Melbourne. The main objects of the Conference were to promote free and active discussion of various aspects of the physical and engineering uses of X-rays, and to provide an opportunity for the presentation of original work being carried out in Australia in these fields. The sixteen papers presented and the discussion on them are summarized; nine of them were concerned with industrial radiography and associated photographic techniques, and the remainder with crystallographic applications.

In opening the proceedings, Dr. F. W. G. WHITE (President of the Victorian Division of the Australian Branch of The Institute of Physics) expressed pleasure at the number (about 120) attending the Conference and the representative nature of the audience. He pointed out the growing interest in Australia in the use of X-rays in various fields, and this, together with the success of a similar type of conference (on Heat Transfer) held in Sydney in 1948, had encouraged the Australian Branch of The Institute of Physics to sponsor arrangements for the present meeting.

During the Conference visits were paid to the Laboratories of the Division of Industrial Chemistry, Commonwealth Scientific and Industrial Research Organization and Defence Research Laboratories, Commonwealth Department of Supply, where a wide range of equipment was on view.

## INDUSTRIAL RADIOGRAPHY

The first of four papers on X-ray equipment for radiography was one entitled "A Brief Review of Industrial X-ray Apparatus" presented by Mr. J. T. CALDWELL (W. Watson & Sons Ltd.). He traced the development of modern equipment through five distinct periods. The gas tube induction coil era lasted from 1895–1915, and the period 1915–1930 saw the widespread application of the hot cathode tube and rectifying valve and the use of alternating current transformers. The third or "shock proofing" period, 1930–1936, was followed by the "supervoltage" period during which the (American) General Electric Company's resonant transformer and van de Graaf electrostatic machines for use up to 2 MeV came into being. During the fifth period, commencing in 1940, devices for the production of high electron speeds without the use of correspondingly high voltages have been developed. These include the betatron, synchrotron and various types of micro-wave accelerators. Of these, the betatron is the first to have reached the commercial stage: it is being produced by the (American) General Electric Company as a 10 MeV unit. This unit has an effective focal spot of about  $1.0 \times 0.14$  mm, and is approximately 6 and 14 times faster than the 2 MeV resonance transformer unit made by this Company when used on 7 and 10 in of steel respectively.

These supervoltage units have very definite uses in heavy industrial fields, but much can be accomplished with units of conventional type. Because of the lag in the development of apparatus specially designed for industrial use it is fortunate that the kilovolt ranges used by the medical profession have been in general conformity with industrial needs; however, much remains to be done on the development of more robust equipment having greater flexibility and, in some cases, portability. An experimental mobile industrial 140-kV unit developed by the Defence Research Laboratories, Commonwealth Department of Supply is shown in Fig. 1(a), while Fig. 1(b) illustrates a further development of this set by Watson Victor Ltd. These units are equipped with 50 ft high-tension shock-proof cables.

During the discussion the author pointed out that all normal types of equipment up to 400 kV are manufactured in Australia with the exception of tubes and valves. No supervoltage units have been installed in this country up to the present.

Mr. Caldwell's passing reference to recent work on the synchrotron was amplified considerably by Mr. W. B. LASICH (Commonwealth Scientific and Industrial Research Organization) in his paper on "The Development of the Synchrotron as an X-ray Generator." The fundamental theory of the betatron was given and the conditions for establishing an equilibrium orbit and for focusing the electrons were derived. The limit to the excitation of the machine is reached when the magnetic saturation of the core causes the electron orbit to collapse. This limitation can be removed by providing a supplementary electric field which oscillates in synchronism with the orbital motion of the electrons. Using this idea, Goward and Barnes<sup>(1)</sup> produced the first synchrotron by over-running a 4 MeV betatron. A brief description of the 18 MeV synchrotron installed in the Physics Department of the University of Melbourne was then given.

The X-ray intensity spectrum is continuous, as is the case with ordinary X-ray tubes. It was shown that the form of the spectrum could be deduced from the interaction of quanta with matter, e.g. the photo-disintegration of deuterium, or by studying pair-production in a cloud chamber. Details were also given of the methods used by Allen-Williams and Appleyard<sup>(2)</sup> for the deter-

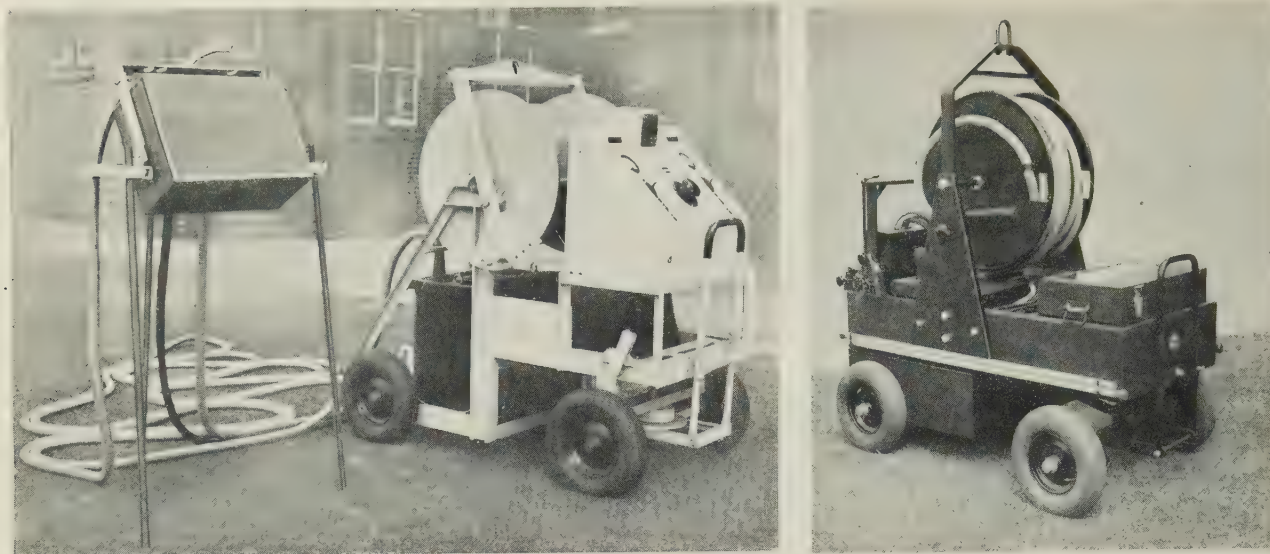


Fig. 1. Mobile industrial 140 kV X-ray unit: (a) (left) As originally produced by the Australian Defence Research Laboratories; (b) (right) As further developed by Watson Victor Ltd.

mination of angular distribution and absorption coefficient in lead of radiation from a 14 MeV machine.

The lecturer then compared the synchrotron and betatron as X-ray generators for industrial use. For the same electron-voltage the synchrotron has the advantage of a much smaller magnet assembly, but the ancillary radio-frequency system adds considerably to the cost of installation and maintenance. Also, whereas the output of a 20 MeV synchrotron weighing about one ton would be of the order of 1 r/min at a focal distance of 1 m, the five-ton betatron will deliver 50 r/min. Although the synchrotron is still in the developmental stages, there is every indication that it may become a useful industrial X-ray generator.

Mr. B. Y. MILLS (Division of Radiophysics, Commonwealth Scientific and Industrial Research Organization) gave a paper on "A Million-Volt Resonant-Cavity X-ray Tube" upon which he had been working. Full details of this investigation are being published elsewhere.<sup>(3)</sup> Briefly, the tube consists of a re-entrant copper cavity resonant in the region of 1200 Mc/s and fed by a 1200 Mc/s magnetron of about 600 kW rated power output. The magnetron is pulsed at 200 c/s by a conventional radar modulator having a 5  $\mu$ sec pulse, and is coupled through a standard wave guide section to a 50  $\Omega$  coaxial line. The coaxial line has a T-junction, one side feeding into the cavity via a coupling loop, and the other into a water load designed to dissipate some of the power and so allow stable operation of the magnetron when the cavity is absorbing large amounts of power. A high-current gun supplied by a separate modulator directs an electron beam across the accelerating gap of the cavity during the period at which the radio-frequency voltage has built up to its maximum value. After crossing the gap these electrons strike a

thin transmission target of gold. The resulting X-rays are confined by a lead shield around the tube to a cone of approximately 15° semi-angle. Provision is made for monitoring the beam current and also the maximum voltage. The cavity is continuously exhausted.

A mean beam current of 70  $\mu$ A at a peak voltage of 1.1 MV has been obtained, whilst at very low currents a voltage of 1.25 MV has been recorded. These figures were obtained when the re-entrant cone tips were in good condition and field emission was very low. They were determined by magnetic analysis of the electron beam, and confirmed by use of an X-ray absorption technique. An attractive feature is that the beam can be focused electromagnetically to produce a small focal spot.

Although a few radiographs have been taken with the apparatus, no attempt has been made to penetrate any very thick sections because the limited capacity of the existing pumping system does not allow of maximum output being obtained for long periods. Mr. Mills estimated, however, that with a beam current of 70  $\mu$ A and using lead screens, a 6-in steel section would require an exposure of about one hour. The pulsed output could be used for stroboscopic examination of moving machinery, etc.

Mr. FARRANDS (Defence Research Laboratories) has recently been actively investigating the possibility of developing this device for use as an industrial X-ray unit. In commenting on Mr. Mill's paper he expressed the opinion that the relatively small output obtained with the apparatus was out of proportion to its complexity. The necessity for continuous evacuation increased its size to the point where it compared unfavourably with existing one and two million volt units, and it appeared that a good deal of maintenance work would be required to keep it operating satisfactorily. In reply,



he author pointed out that the apparatus was still in the experimental stages, but would doubtless be improved by subsequent developments. There would appear to be no fundamental reason why the tube should not be cooled off and the actual X-ray head would then be extremely compact.

The last paper on X-ray equipment was one by Mr. R. G. ACKLAND (Defence Research Laboratories) on "Flash Radiography." After briefly describing "millisecond" techniques in which normal hot-cathode-type tubes can be used, the lecturer dealt in some detail with the evolution of "microsecond" radiography, and the problems involved in the development of the special equipment required for this work.

To produce reasonable results in single microsecond exposures, tube currents of the order of 1000 A are required. Condenser type power supplies can provide these large momentary currents quite satisfactorily. Single units can be used up to 100 kV, but multi-stage Marx-type generators are usually more convenient for higher voltages. When taking microsecond exposures, it is obvious that careful attention must be given to the design of the apparatus used to initiate the discharge of the power supply.

The first X-ray tubes to handle these very large currents utilized a mercury-pool-type cathode, in which, by means of a special starting electrode, a hot-spot capable of emitting sufficient electrons was produced. This type of tube did not prove to be suitable for practical use. However, in 1940, Slack and Ehrke<sup>(4)</sup> produced a cold-cathode tube which proved to be satisfactory and which is now manufactured commercially by the Westinghouse Electric Corporation. In this tube an auxiliary starting electrode placed within the cathode cup produces fields sufficiently intense to result in cold emission of electrons from the cathode surface. In the Westinghouse flash unit this tube operates at voltages up to 350 kV, and passes currents of the order of 1000 A. By using a separate power supply for the starting electrode, Clarke<sup>(5)</sup> claims that he has passed 30,000 A through the tube in  $0.1 \mu\text{sec}$ . After dealing with methods used for accurately gauging the operating characteristics of flash equipment, the author briefly described the units made by the Westinghouse Electric Corporation, the Armament Research Establishment at Siemens Halske, and quoted some examples of the uses which are made of this technique.

Dr. C. E. EDDY (Commonwealth X-ray and Radium Laboratory) presented a paper entitled "Some Limitations in Interpreting Radiographs." Attention was first paid to the many factors which can influence the resolving power of a radiograph, including the effects of various geometrical arrangements of source, work and film; the use of intensifying screens and films of different grain sizes; and radiation scattered both from external objects and from within the work itself. The importance of correct selection of tube voltage was emphasized, and, where a wide range of thickness or composition is

encountered, the advantages to be gained by taking two or more radiographs at different voltages were pointed out. Tube current and exposure times must also be adjusted to give the most suitable range of densities in the film.

As the final examination of a radiograph is made by the eye, conditions of viewing should be such that the eye functions most effectively. High intensities of illumination and high densities can be used with advantage. It has been found that there can be appreciable difference between the efficiencies of individual observers; quite wide variations in interpretation can occur, especially if a subjective rather than an objective standard is used as a criterion, and these variations are increased if the radiographs being inspected vary in quality. In order that objective measurement should supersede subjective judgment, it is highly desirable that step-wedges and other similar devices should be used.

Many of the points mentioned briefly by Dr. Eddy in his paper were covered more fully by Messrs. W. F. HEMMY and I. J. FERRIS (Defence Research Laboratories) in their papers on "Problems Associated with Scattered Radiation" and "Contrast and Definition in Industrial Radiography" respectively.

Mr. Hemmy pointed out that any objects in the X-ray beam will serve as sources of scattered radiation, the intensity and penetrating power of which, in general, increase with increasing tube voltage and material thickness. Lead diaphragms at the tube port can be used to limit an unnecessarily wide beam, or the scattering objects may be shielded by lead sheet. The most effective method of reducing scatter from objects, including the work, that cannot be shielded from direct radiation is by the use of lead filters, either at the tube port or immediately in front of the film cassette. In the first case the softer radiation of the primary beam, which, owing to its low penetrating power would contribute mainly scatter to the radiograph, is scattered away from the film. In the second, the filter absorbs the scattered radiation generated in the specimen itself to a greater degree than the image-forming radiation. The use of metal intensifying screens in contact with the film has the advantage that not only is soft scattered radiation filtered out, but also exposure times are reduced because of the highly efficient image-forming electrons generated in the screen. The intensification factor of these screens is low (two to five), but it increases with diminishing wavelength of the primary beam and, unlike fluorescent screens, is not subject to failure of the reciprocity law.

To obtain good images of the boundaries of a specimen use must be made of lead sheet, shot, or solution blocking, otherwise the relatively intense beam reaching the film past the edge of the specimen can generate considerable scatter in the film itself giving an effect similar to photographic halation.

Mr. Ferris drew attention to the fact that two of the more important factors governing "radiographic sensitivity" are radiographic contrast and image definition,

Contrast is the difference in film density between the image of a defect and its surroundings, and for a given sample may be varied by changing X-ray tube voltage, by selecting films of different characteristics and by the use of techniques that will reduce the effects of scatter. Image definition is the sharpness of image outline and is dependent on the geometry of the set-up and the grain size of the film or film-screen combination. Most factors which improve contrast and definition increase the length of exposure, so that the radiographer must select optimum conditions of contrast and definition to give the best sensitivity consistent with a reasonable exposure time.

The use of penetrameters for measuring sensitivity was illustrated, and the uses and limitations of the American and British flat types, which essentially measure contrast, and the Continental types using groups of fine wires, which measure image definition rather than contrast, were discussed.

In the discussion which followed these three papers the importance of adequate and uniform illumination for viewing films was stressed. The part played by photo-electrons from metal screens in producing intensification of the image and the use of screens for making "electron" radiographs were mentioned.

Dr. N. B. LEWIS (Research Laboratories, Kodak (Australasia) Pty. Ltd.), in his paper on the "Processing of Radiographic Materials," dealt in some detail with the nature of the latent image, its mode of formation in the emulsion, and the mechanism of development. He then considered the difficulties involved in ensuring uniform development, a most important requirement for the production of satisfactory radiographs, especially if they are to be used for the quantitative estimation of defects. Uniformity of development is obtained only if the composition of the developer in contact with the film is uniform over the whole surface. Normal diffusion processes, which tend to remove local variations in developer concentration, are too slow, so that some method of agitation is necessary. Methods recommended by the American Standards Association (Z38.83) and by the British Standards Institution were described, and reference was made to more elaborate methods such as that described by Morgan and van Allen.<sup>(6)</sup> While adequate agitation is important, excessive agitation can produce uneven development by setting up flow patterns in the developer.

In radiography, reproducibility of development is not, in general, as important as uniform development of a specific film. If reproducibility is required, care must be taken to see that the same amount of agitation exists for each film and that the developer composition remains constant. The uses of "replenishers" and replenishment techniques were described.

Finally, since the important quantity in radiographic work is contrast, the effect of different developing techniques on contrast was also discussed. The occurrence of fog affects contrast adversely, so that the technique

of fogging after exposure to increase the effective speed is not advisable; the production of chemical fog by excessive development should also be avoided.

Of interest to all concerned with X-ray work was a paper by Mr. D. J. STEVENS (Commonwealth X-ray and Radium Laboratory) on "Protection in Industrial Radiography." He discussed briefly the types of injuries which may be produced by X-rays and gamma-rays, emphasizing that the effects of exposure are cumulative. Reference was made to the activities of various Protection Committees,<sup>(7)</sup> and their work in establishing the "tolerance dose." Slides, many of them in colour, graphically illustrated the disastrous injuries which can result from ignorance of the dangers involved or carelessness. Tables showing the biological effects resulting from various dosages and the values of X-ray output from various types of industrial X-ray apparatus served further to emphasize that extreme care must be taken in providing suitable protection and automatic safety devices. Not too much faith should be placed in the safety of so-called "protected" tube heads, owing to the various definitions of the term. Furthermore, danger arises from the radiation scattered by the object being radiographed. Finally, the desirability of carefully checking each installation for satisfactory protection and of using personnel monitoring systems was stressed.

During the discussion reference was made to the dangers inherent in the unrestricted use in shoe shops of X-ray fluoroscopes. The use of fluorescent screens for checking the alinement of X-ray diffraction cameras was mentioned as a possible source of danger, unless protected by lead glass.

#### X-RAY ANALYSIS

The first paper dealing with the crystallographic uses of X-rays was given by Dr. R. I. GARROD (Defence Research Laboratories) on "Considerations in the Design of X-ray Diffraction Equipment," and was limited to X-ray tubes and powder cameras. After a preliminary review of the relative virtues of sealed-off and demountable type tubes, some general design principles for optimum performance of demountable type crystallographic tubes were discussed, and a description given of two types of tubes which have been designed and constructed at the Defence Research Laboratories.<sup>(8)</sup> One of these tubes is of the four-windowed vertical type, employing a single filament giving a line focus, and incorporating an externally adjustable filament-focusing device. The advantages of employing a line focus in four-windowed tubes were pointed out.<sup>(9)</sup> The second tube is of the horizontal type, but also includes the virtue of the vertical type tube in having the high-tension generator below table level.

The second part of the paper dealt with general design principles for powder cameras, and described constructional details of an 11.46 cm diameter camera which has been constructed recently to the design of Dr. Garrod, Dr. Wood (University of Melbourne) and



Dr. Boas (Division of Tribo-physics, Commonwealth Scientific and Industrial Research Organization). A view of the main parts of the camera is given in Fig. 2. The camera is suitable for use with either vertical or horizontal type tubes and consists of a main body *A*, which houses a light-tight cassette *B*, containing the film holder, collimator and beam-trap. A novel means is incorporated for clamping the film accurately against a given cylindrical surface, at the same time permitting easy and convenient loading in the dark room. The specimen-entering device utilizes polar co-ordinate adjustment and may be performed in a single operation by an external control knob.

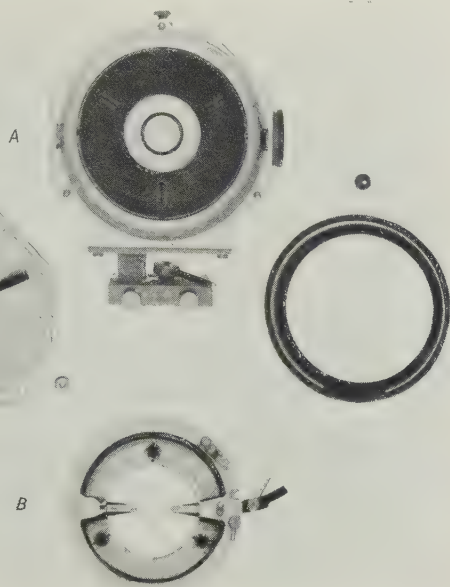


Fig. 2. Component parts of 11.46 cm diameter powder camera

The second paper in this session was read by Dr. A. MCL. MATHIESON (Division of Industrial Chemistry, Commonwealth Scientific and Industrial Research Organization) on "High-power Generators for X-ray Diffraction." Exposure times for recording diffraction patterns photographically are governed ultimately by the maximum permissible specific loading on the target of the X-ray tube. In consequence considerable attention has been devoted in recent years to the development of high-power X-ray generators<sup>(10,11)</sup> usually incorporating rotation, oscillation or gyration of the anode to increase the maximum permissible power input. After reviewing previously designed tubes of this type, the author described a high-power X-ray generator which is at present being constructed in the laboratories of the Division. The gyrating anode principle is employed, thus simplifying water cooling of the target. In shape the target is a domed cylinder inside a vacuum-tight magnesium alloy casing and supported on a shaft which carries the cooling water. The shaft passes through a universal ball-joint

and fits at its lower end into a socket in the drive disk. The diameter of the anode is  $13\frac{1}{2}$  cm and of the focal path  $10\frac{1}{2}$  cm, with an angular range for gyration of  $10^\circ$ . It is proposed to use a rubber bellows as the vacuum seal for transmitting this large angular motion. The cooling water after passing through the X-ray tube is cycled through a cooling radiator and pumped back to the anode. The filament is a stout tungsten rod set in an equipotential plate at a distance of about 8 cm from the anode. To produce a sharp focal spot, a magnetic lens is incorporated to produce a slightly reduced focal image of the filament on the anode. Three-phase supply with full-wave rectification is used for the high-tension voltage for the unit, the h.t. being variable from zero to 50 kV. It is anticipated that the tube will operate at up to 50 kV with a beam current of 200 mA. Dr. Mathieson concluded by outlining some of the applications for which it is intended to use the generator. The principal use will be the study of the structure of proteins. In combination with a multiple camera, a series of exposures will be recorded on a single film from specimens under varying conditions of temperature, humidity and tension. Again, the high intensity source from the generator will permit the employment of monochromators for studying particle size in low-angle-scatter work, and for recording diffuse reflexions due to thermal motion, without an unduly long exposure time.

The final day of the Conference was devoted to applications of X-ray crystallographic methods to problems in pure and industrial research. The session was opened by Dr. W. A. WOOD (Baillieu Laboratory, University of Melbourne) with a review of the past history of X-ray work in Great Britain and of his own contributions in this field. After discussing several typical examples of early applications in several fields, the author went on to review more recent work, mainly in the field of metal physics, in which he had been directly engaged. Broadly speaking, the investigations could be divided into four main classes. First, there was the study of distorted structures, typical examples being electro-deposits,<sup>(12)</sup> nitrided steels<sup>(13)</sup> and magnetic materials. Secondly, there were the strains produced in the atomic lattice associated with the application of stresses to polycrystalline metals, and the discovery of the existence of permanent residual lattice strains after removal of an applied stress in excess of the yield. The origin of the internal stress system, created during the process of plastic deformation and responsible for these residual lattice strains, is still a matter for speculation.<sup>(14)</sup> Thirdly, there were investigations into the crystalline texture of materials and the changes produced by deformation under varying conditions.<sup>(15)</sup> Reference was also made to the changes occurring in mica at elevated temperatures.<sup>(16)</sup> Lastly there were the very interesting structural effects associated with fatigue in metals under conditions of static and alternating loads.<sup>(17)</sup> The speaker concluded with a brief reference to work which he had commenced at the National

Physical Laboratory and is now carrying on in Australia on the behaviour of metals at elevated temperatures under creep conditions,<sup>(18)</sup> and pointed out that the extent of disorientation of the grains as a result of creep stresses depends markedly upon the rate of strain and the temperature.

In the discussion which followed, Mr. F. E. OCKENDEN (W. Watson & Sons) raised a point regarding the behaviour of hair-springs in precision electrical indicating instruments. It is common practice to use springs in which the torsion necessary to cause yield is enormously greater than that to which the movement is normally subjected. Yet if the movement is twisted through 90° and left for some time, the springs acquire a permanent set which gradually disappears with time. In reply, Dr. WOOD suggested the behaviour was one associated with elastic after-effect and possibly due to viscous flow at the grain boundaries. Dr. BOAS (Commonwealth Scientific and Industrial Research Organization) supported this view and pointed out that in single crystals such effects are generally very much reduced.

The second contribution to the session was a joint paper by Messrs. J. H. AULD and K. A. GROSS (Defence Research Laboratories) on "Applications of X-ray Analysis to Industrial Problems." This paper was essentially a review, with reference to typical examples, of applications to industrial problems in chemistry and metallurgy. After briefly outlining the fundamental principles of X-ray techniques, Mr. Gross illustrated the applicability of the powder camera method to the qualitative and semi-quantitative analysis of crystalline materials, and pointed out the limitations at present existing in the A.S.T.M. index. References were then made to the accurate determination of lattice spacings by the back-reflexion technique, the uses and applications of low-angle-scatter methods, and the determination of particle size in the sub-microscopic field. Dealing with metallurgical applications, Mr. Auld first described the principles and techniques involved in detecting the presence of preferred orientation in polycrystalline metals, and the effects of such orientation on the processes of cold working, recrystallization after cold work and the electrodeposition of metals. Mention was then made of applications to the study of the effects of heat treatment of alloys, with special reference to recrystallization, quenching and tempering, age hardening and heat treatment of magnet steels. A third important field includes applications of chemical analysis by X-ray methods, such as the identification of phases, corrosion products, scales, etc. Lastly, the speaker discussed the applications of the method and its limitations to the determination of internal stresses in metals.

In the subsequent discussion, Mr. NORRISH (Division of Soils, Commonwealth Scientific and Industrial Research Organization) stated that in a number of cases he had found the data in the A.S.T.M. index unreliable. He also discussed the difficulties of preparing, in certain cases, specimens for powder camera work without some

degree of orientation. Dr. ADCOCK (Broken Hill Pty. Ltd.) remarked on the change of line width with the quality of X-radiation employed, and asked if the use of monochromatic radiation in such cases was important. In reply, Mr. GROSS outlined methods for obtaining monochromatic radiation, and Dr. GARROD discussed briefly present views on the causes of line broadening from cold-worked metals, and pointed out the desirability of using monochromatic radiation in such investigations.

The third paper was given by Dr. R. I. GARROD (Defence Research Laboratories) on "Internal Stresses in Polycrystalline Metals." When stresses are applied to a polycrystalline metal aggregate the individual

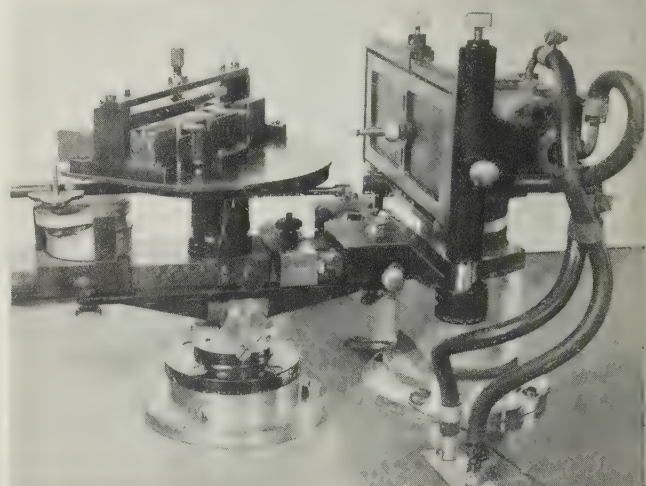


Fig. 3. Equipment for determining stress v. lattice strain relationships in metals

crystals deform, and by using precision back-reflexion X-ray techniques it is possible to determine the relationships between the applied stress and the resulting lattice strain in certain crystallographic directions. Such stress-strain relationships will in general differ from the results obtained for the same material by standard engineering methods, owing to the selective nature of the X-ray method and the anisotropic elastic behaviour of most metals.<sup>(14)</sup> The author described some preliminary results of work of this type which he is at present carrying out on a low carbon steel. Annealed specimens of steel were placed in a precision self-aligning four-point bending fixture (Fig. 3) and subjected to progressive uni-axial loading, X-ray photographs being taken at normal and oblique incidence for each load. By using different X-radiations, a series of applied stress versus lattice strain relationships was determined for different families of planes, for both tensile and compressive stresses. The following results were obtained. (a) Even after the application and removal of a stress well below the nominal yield point for the material, a small but detectable residual lattice strain was observed. (b) Within the nominal elastic range, the lattice behaved elastically



to any value of previously applied stress, but if this were exceeded the strain was no longer proportional to the stress, and a fresh residual lattice strain was obtained on unloading. (c) If after straining one side of a specimen under tensile load by bending (still below the nominal yield stress) and the same side was then subjected to compressive stress, the material exhibited a appreciable Bauschinger effect, i.e. the subsequent elastic range in compression was considerably reduced. The values of lattice strain/unit stress obtained experimentally were compared with theoretical values computed on the assumptions (i) that the strains in all the individual crystals are the same, and (ii) that the stress remains constant throughout the aggregate. Comparative values are given in the table, and it will be seen that, in general agreement with previously reported work of this type,<sup>(19)</sup> neither of the two limiting cases is valid in practice. This work will be reported in more detail elsewhere.

Table 1. Lattice strain/Stress values in  $\text{mm}^2/\text{kg} \times 10^{-5}$  for different families of (hkl) planes

(hkl)	$\alpha$	Theoretical values		Experimental	
		Constant strain	Constant stress	Tension	Compression
(310)	81°	-1.08	-2.02	-1.75	+1.75
	41°	+1.94	+2.66	+2.68	-2.67
(211)	79°	-1.01	-1.07	-1.13	
	44°	+1.65	+1.69	+1.67	
Average					
for (651)	77°	-0.93	-1.18	-1.05	
and (732)	42°	+1.84	+2.03	+1.93	

( $\alpha$  is the angle between the reflecting normals and the stress direction.)

The author then went on to describe some complementary work also being carried out on residual lattice strains present in the material after it has been subjected to homogeneous tensile loads in an Amsler machine. He has found that after removal of an applied stress, residual lattice strains exist for various families of planes in the lattice, and that, in particular, the strains for those planes in which the 220 reflecting normals are approximately perpendicular to the direction of previously applied stress, change in sign as the stress is increased.<sup>(20)</sup> Dr. Garrod stated that it is difficult to reconcile these results with a theory put forward recently by Greenough.<sup>(21)</sup> In the subsequent discussion, points were raised regarding the reasons why values for Poisson's ratio (assuming isotropic behaviour) obtained by X-ray methods are larger than the engineering values, and also the comparative nature of the information obtained from locked-up stresses by X-ray and photo-elastic methods.

The afternoon session was opened by a paper from Mr. K. NORRISH (Division of Soils, Commonwealth Scientific and Industrial Research Organization) on "X-ray Analysis applied to the Layer Lattice Silicate Minerals." In his opening remarks, the speaker pointed

out that his interest in this subject was twofold. First, in extending the range of available X-ray data for the identification and differentiation of layer lattice silicates, and, secondly, in the actual crystal structures of minerals of this class and the types of lattice defects which are found to exist. Since most of the minerals occur as very fine particles, he has employed the powder technique almost exclusively, and because of the nature of the work has found it necessary to design his powder camera to record diffractions from interplanar spacings up to at least 20 Å. Because of the poor resolution in the region of the basal spacings of clays, the diffraction lines often overlap on ordinary powder photographs. However, the resolution and intensity of the 00l lines can be improved by preparing the sample in the form of a thin flake, approximately 0.5 mm wide, and placed in the camera so that the plane of the flake is parallel to the incident X-ray beam. In connexion with the crystal structure of layer lattice silicates the speaker pointed out that the layers of these minerals have one of two basic structures (which leads to similarities in their diffraction patterns) although the methods of stacking the layers differ for most of them.<sup>(22)</sup> The two types of layer structure, termed single and double layer types, differ only in that the latter has an extra sheet of tetrahedrally co-ordinated silicon ions. This means that the *a* and *b* axes of the unit cells of all layer lattice silicates are much the same. Their *c* axes, however, vary considerably. In some double layer type structures in which the layers have a net negative charge (such as montmorillonite and chlorite) it has been found that particle size and hardness, and the way in which the layers pack together, are dependent on the layer charge. Whilst some minerals such as kaolinite have an ordered structure, others such as halloysite have random arrangements in which the layers are parallel and equally spaced but randomly oriented with respect to one another. In consequence the *hk0* reflexions from this latter type of structure are broad and asymmetrical.<sup>(23)</sup> Finally, the speaker referred to the ability of some silicates to form mixed layer crystals. Because of their similarity, layers of one mineral can sometimes be stacked at random with those of another mineral. The diffraction patterns of minerals of this type have weak and diffuse 00l reflexions which, furthermore, do not precisely obey Bragg's law.<sup>(24)</sup>

In the discussion which followed, Dr. GARROD enquired the reason why in some of the patterns illustrated the 00l lines appeared to consist of two semicircles of different diameter. Mr. NORRISH suggested this was due to variation in packing of the crystals in the flake of the specimen. Dr. GARROD also discussed powder camera design, and pointed out that for general flexibility of application the requirements of reasonable exposure times and recording high and low  $\theta$  values were somewhat conflicting. Usually, therefore, a compromise is made between these two requirements in a general purpose powder camera.

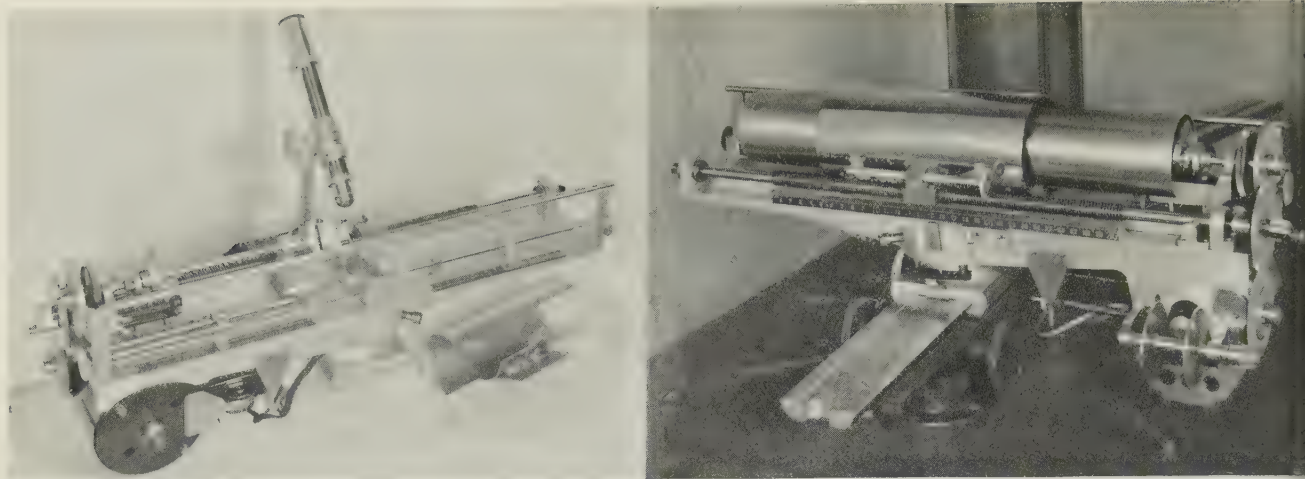


Fig. 4. Weissenberg equi-inclination goniometer: (left) Front view, with shield in foreground; (right) Rear view

The final paper of the Conference was given by Dr. A. McL. MATHIESON (Division of Industrial Chemistry, Commonwealth Scientific and Industrial Research Organization) on "Single Crystal Methods in Structural Analysis." After reviewing the technique involved in obtaining diffraction data from a single crystal, and the indexing of reflexions by using the reciprocal lattice concept, the author went on to describe an equi-inclination Weissenberg goniometer which has been designed and constructed in his laboratories. This instrument will be described in detail elsewhere,<sup>(25)</sup> but an illustration of it is given in Fig. 4. The collimator is rigidly attached to the base and is thus fixed with respect to the incident beam. The frame, containing crystal and film holders and their associated drives, can be rotated on the base (up to an angle of  $30^\circ$ ) so that the incident and diffracted beams are equally inclined to the rotation axis of the crystal. By this means the reflexions can be indexed conveniently and their intensities obtained. The next step is to synthesize the experimental data to yield contour maps, from which atomic parameters can be measured and bond lengths computed. Dr. Mathieson discussed the various methods available for carrying out this process, and pointed out that the study of structure by X-ray analysis differs from other methods in that a complete agreement of observed and calculated data is necessary. No deduction is possible from use of part of the available information and in consequence the results are correspondingly more precise. Discussion on this paper between Dr. Mathieson and Mr. McCONNELL (Sydney Technical College) was centred mainly on the accuracy necessary in determining diffracted intensities, for application of the Patterson-Harker series in contour plots.

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R. I. GARROD  
J. F. RICHARDSON

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## ORIGINAL CONTRIBUTIONS

## Method for Measuring the Efficiency of Getters at Low Pressures

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[Paper first received 4 April and in final form 28 April, 1950]

A method is described for measuring the rate at which gas is taken up by a getter at low pressures. The measurement is based on ascertaining the drop in pressure along a narrow tube through which the gas to be absorbed by the getter is passed. The pressures on either side of the tube are measured by means of ionization gauges. The values found for the rate of gettering (of air) vary between  $10 \text{ cm}^3 \text{ sec}^{-1}$  for magnesium and  $1500 \text{ cm}^3 \text{ sec}^{-1}$  for a thorium getter. The values depend on the position of the getter in the valve and increase with increasing proximity between the getter and the region in the valve in which the gas molecules are exposed to the impact of electrons.

## 1. INTRODUCTION

The efficiency of getter materials which are used for improving the vacuum in electronic valves is normally measured by determining the total quantity of gas which can be absorbed by the getter. Such quantities, for instance, have been given by Reimann<sup>(1)</sup> and by Clarke and Slack<sup>(2)</sup> who measured in the pressure range between  $10^{-3}$  and  $10^{-2}$  mm mercury. The quantities found were between 10 and 100 l of gas if related to a pressure of  $1 \mu$ . The knowledge of these quantities is useful if—as in commercial pumping—pressures between  $10^{-3}$  and  $10^{-2}$  mm are actually used on the pump. The possibility of reducing these pressures to a working pressure of about  $10^{-5}$  mm in the valve can then be assessed.

The quoted quantities of gas, however, become rather relevant if the possibilities of obtaining an extremely low pressure in the valve are to be discussed. Let us assume that a pressure of  $10^{-5}$  mm has been established in the valve by some means, either by proper pumping with a diffusion pump or by means of a getter supplied to the valve for this particular purpose. The quantity of free gas in the volume of the valve, assuming a volume of  $50 \text{ cm}^3$ , then amounts to only  $5 \times 10^{-4} \mu\text{l}$ . This quantity is negligibly small compared with the total quantities of  $10 \mu\text{l}$  or more which can be adsorbed by a getter. Such a small quantity can be easily adsorbed not only by a getter, but by any surface in the valve which has previously been freed from gas sufficiently. The real problem which arises is no longer a problem of adsorbing quantities of gas in general, but of directing these quantities from other parts of the valve to the getter in order to locate them there permanently. It is necessary, for instance, to avoid some of the gas staying on or at the cathode where it would damage the performance of the valve during further operation. The factor which is determining these processes is the rate at which the gas is taken up by the getter, and it is this rate of gettering with which this investigation will be concerned.

Before discussing the method which has been developed

for measuring the rate of gettering, an example may be given which shows that the rate of gettering must be expected to be different with different getters. This example was obtained when some experimental tetrodes (for their design, see Fig. 3a) were supplied with different getters (barium, magnesium, zirconium and thorium, for details see Section 5). These valves were pumped on a diffusion pump in exactly the same manner and sealed off at a manifold pressure of  $10^{-5}$  mm. After sealing-off, these valves were run with 5 mA anode current and 200 V on the anode, whilst the pressure in the valves was measured by collecting the positive ions

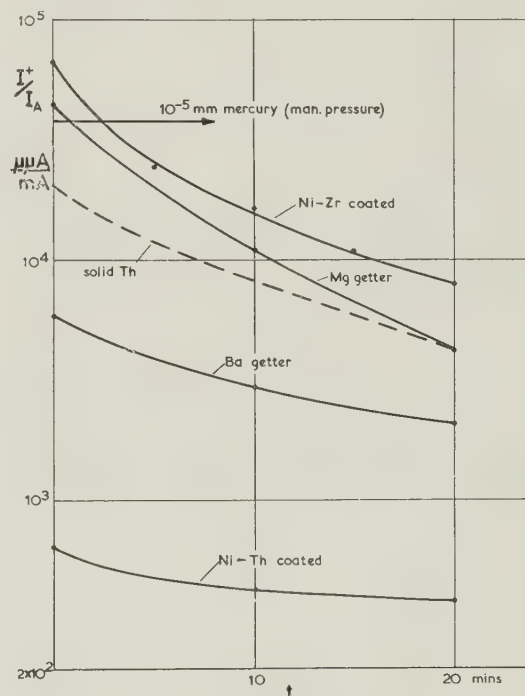


Fig. 1. Ratio  $I^+/I_A$  of positive ion current and anode current (vacuum factor) in valves with different getters, measured as a function of time after sealing-off

produced in the valve on special collector wires ( $C$  in Fig. 3). The ratio between the positive ion current  $I^+$  and the anode current  $I_A$  is plotted in Fig. 1 as a function of time. The figure shows that there was a wide diversity between the ion currents and therefore between the pressures existing in the valves with different getters. The different efficiency of these getters at the pressures concerned is thus demonstrated.

## 2. FUNDAMENTALS OF NEW METHOD

The method which has been used for determining the rate of gettering is based on Knudsen's laws for the flow of gas at low pressures.<sup>(3)</sup> Fig. 2 shows a sketch

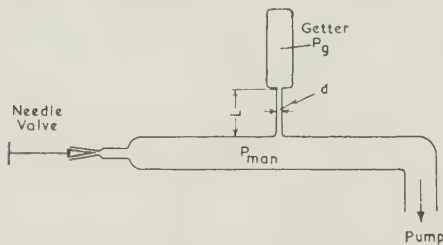


Fig. 2. Experimental arrangement for measuring the rate of gettering

of the experimental layout. The getter to be examined is located in a glass bulb of about 50 cm<sup>3</sup> volume and connected to the manifold of the pump by a pumping stem of a small diameter (the values actually used are  $d = 2$  mm internal diameter and  $L = 40$  mm length). The manifold can be pumped to a minimum pressure of  $10^{-7}$  mm by means of a diffusion pump. Furthermore, a needle valve is connected to the manifold, and by using this valve the pressure  $p_{man}$  in the manifold can be adjusted to any higher value which is required for the measurement.

As the getter in the glass bulb continuously adsorbs gas the pressure  $p_g$  in the bulb will be smaller than  $p_{man}$ . The difference between these two pressures is given by Knudsen's formula,

$$(p_{man} - p_g)F = p_g \frac{dV}{dt} \quad (1)$$

where  $F$  denotes the conductivity of the pumping stem and  $dV/dt$  the volume of gas flowing through the pumping stem and adsorbed by the getter per unit of time. This latter quantity is identical with the rate of gettering  $G$ , and therefore

$$G = F \frac{p_{man} - p_g}{p_g} \quad (2)$$

or, if

$$p_{man} \gg p_g$$

$$G \simeq \frac{p_{man}}{p_g} F \quad (3)$$

The rate of gettering  $G$  can be determined from equation (2) or (3) if the 2 pressures  $p_{man}$  and  $p_g$  are measured and if the conductivity  $F$  of the pumping

stem is known. The conductivity  $F$  is normally given in cm<sup>3</sup> sec<sup>-1</sup>, and  $G$  will therefore be obtained in the same unit, referring to the pressure  $p_g$  existing above the getter. The conductivity  $F$  can either be determined experimentally or be calculated from Knudsen's formulae.<sup>(3)</sup> The value obtained from these formulae for  $d = 2$  mm and  $L = 40$  mm is  $F = 20$  cm<sup>3</sup> sec<sup>-1</sup> (for air). The values obtained experimentally will be given under 4.

## 3. MEASUREMENT OF THE PRESSURE

For measuring the pressure  $p_{man}$  in the manifold a commercial ionization gauge was used which allowed measurements in the range between  $10^{-7}$  and  $10^{-3}$  mm. The gauge for measuring the pressure above the getter had to allow measurements below the limit of  $10^{-7}$  mm and had to be combined with the carriers of the different getters. A special gauge had therefore to be designed: it was obtained by modifying experimental tetrodes which had been used for other investigations. Fig. 3 shows the two types of gauges which were used. The cathode providing the electrons was an indirectly heated oxide cathode. The positive gas ions formed by those electrons were collected on collector wires  $C$  which were located at the position of the suppressor grid in normal pentodes. These wires protruded into the valve assembly from the end opposite to the glass pinch carrying this assembly; they were supported by a second pinch sealed into the glass bulb at the opposite end. Owing to this arrangement a high insulation between the collectors and the other parts of the assembly could be obtained. Furthermore, owing to the wide distance between the collectors and the cathode, the collectors remained fairly cool during the operation of the valve, and there was consequently no measurable thermal emission from these wires which could interfere with the measurements.

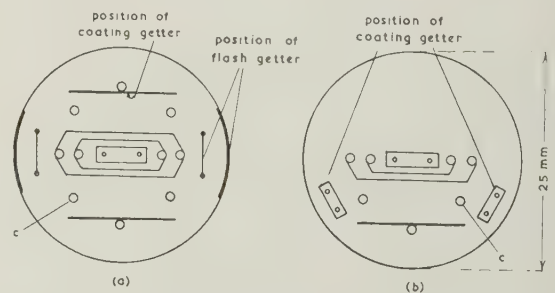


Fig. 3. Cross-sections through the valves used for the measurements

The normal conditions for operating this ionization gauge were: screen grid voltage  $V_{G2} = 25$  V, anode voltage  $V_A = 200$  V, anode current  $I_A = 10$  mA, collector voltage  $V_C = -15$  V. The cathode temperature was 1250° K, it was deliberately kept high in order to prevent the cathode from taking up gas and competing with the getter. The collector current  $I_c$  was measured.



y means of a galvanometer which had a sensitivity of  $4 \mu\text{A}/\text{mm}$  with a scale distance of 2 m.

The pressure  $p_g$  existing in the ionization gauge is obtained from the relationship:

$$K = I_{\text{col}}/I_A = Cp_g \quad (4)$$

where  $K$  is called the vacuum factor, and  $C$  is the calibration constant of the gauge. The calibration constant  $C$  was determined by comparing the ionization gauge shown in Fig. 3a with the commercial gauges used for measuring the pressure in the manifold. In order to avoid errors, due to the drop of pressure along the tubes connecting the gauges, the gauge to be calibrated was enclosed between 2 of the commercial gauges, and the diameter of the connecting tubes was kept wide (inside diameter  $\approx 10$  mm). The readings of the two gauges, used for the calibration, differed by about 30% on the average. The mean value between these two readings was used for plotting the calibration curve shown in Fig. 4 (vacuum factor  $K$  in  $\mu\text{A}/\text{mA}$  against pressure).

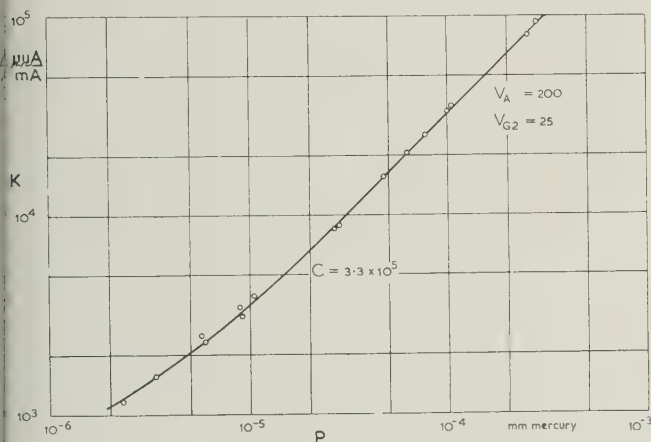


Fig. 4. Calibration curve of the ionization gauge used for measuring the pressure in the valves

As shown in two papers by Metson<sup>(4, 5)</sup> the simple relationship of equation (4) is only valid for higher pressures. When, with decreasing pressure, the ion current becomes very small, another phenomenon must be taken into account, namely the emission of photo-electrons from the collector surface which are produced by soft X-rays coming from the anode. These photo-electrons emitted from the collector cannot be distinguished from the gas ions collected there; only the total current, due to photo-electrons plus ions, can be measured. As the photo-electric current is independent of pressure, the vacuum factor  $K$ , as defined by the ratio of collector current and anode current, will approach a constant level, when the pressure is decreased more and more. If this level which is due to X-ray radiation is denoted by  $K_{\text{rad}}$  equation (4) has to be replaced by

$$\frac{I_{\text{col}}}{I_A} = K \quad (5)$$

$$K - K_{\text{rad}} = Cp_g$$

The existence of the radiation level is seen in the calibration curve of Fig. 4 by a deviation from linearity below a pressure of  $10^{-5}$  mm. At these low pressures a quantitative measurement of the pressure is only possible, if the radiation level is known. In the experiments described here the radiation level could be ascertained if the pressure in the manifold was lowered sufficiently. Fig. 5 shows the values of the vacuum factor  $K$ , as obtained for a particular getter (thorium getter, see Section 5), plotted against the pressure  $p_{\text{man}}$  in the manifold. Let us first consider the lower of the two curves, denoted by  $K_{25}$  (the subscript refers to the screen grid voltage of 25 V). If the manifold pressure  $p_{\text{man}}$  is sufficiently low the vacuum factor becomes practically independent of  $p_{\text{man}}$ . The pressure in the valve with the getter then is so low that the ion current becomes negligibly small compared with the photo-electric current produced by the X-rays. The radiation level can therefore be obtained from the horizontal part of the curve concerned.

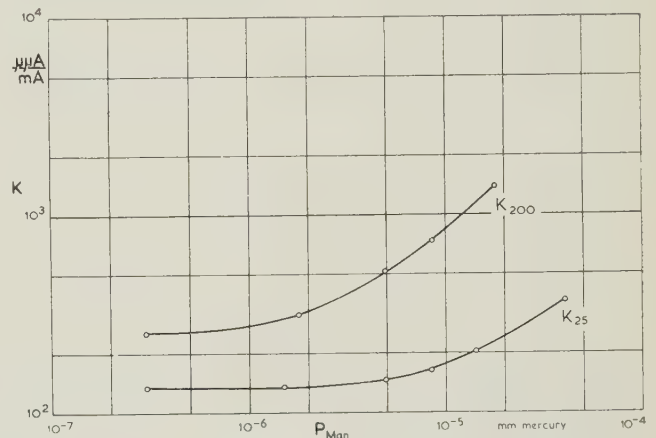


Fig. 5. Vacuum factor  $K$  as a function of manifold pressure  $p_{\text{man}}$  for a thorium getter (Thorium deposited on anodes)

$K_{25} : V_{G2} = 25 \text{ V}$

$K_{200} : V_{G2} = 200 \text{ V}$

The sensitivity of the ionization gauge used could be increased by applying 200 V to the screen grid instead of the normal 25. A curve of the vacuum factor  $K_{200}$  measured in this way is also shown in Fig. 5. In this case, however, the determination of the radiation level was made more difficult, because a horizontal part of the curve could not be obtained owing to the fact that the ion currents were no longer negligibly small even at the lowest pressures. Hence the gain in sensitivity, produced by the increased screen grid voltage could not be used with advantage. The vacuum factor  $K_{200}$  was therefore employed only in a few cases, where it could be shown that the results obtained agreed with those obtained by using  $K_{25}$ .

#### 4. CALIBRATION OF STEM TUBE

For a quantitative evaluation of the gettering rate  $G$  according to equation (3) the conductivity  $F$  of the

stem tube, connecting the experimental valve with the manifold, must be known. The conductivity  $F$  can be determined experimentally, if a known volume  $V$  is evacuated through the stem tube concerned. The process of evacuation then is governed by the equation

$$-V \frac{dp}{dt} = Fp \quad (6)$$

which can be solved by an exponential function  $p = \exp(t)$  the exponent of which contains the unknown conductivity  $F$ .  $F$  can therefore be determined by plotting  $\log p$  against time and ascertaining the slope of the straight line obtained.

Two such determinations were undertaken in which an ionization gauge was used for measuring the pressure. As the ionization gauge itself acts as a pump which evacuates the volume connected to the pumping stem, the pumping speed of the gauge had to be measured in a preliminary experiment. The average pumping speed found was about  $5 \text{ cm}^3 \text{ sec}^{-1}$ . This value was used for correcting the value measured for the conductance  $F$  of the pumping stem. The final result was an average value  $F = 18.5 \text{ cm}^3 \text{ sec}^{-1}$ , which compares fairly well with the theoretical value of  $20 \text{ cm}^3 \text{ sec}^{-1}$  (see Section 2).

## 5. TYPES OF GETTERS EXAMINED AND PROCESSING OF EXPERIMENTAL VALVES

There are two main types of getters, the flash getters, such as barium, which are flashed on to the glass wall of the evacuated valve, and the coating getters, as, for instance, zirconium, which are deposited on metal components of the valve, before these components are built into the valve. Two getters out of each of these groups were investigated, and they were located in the valve and processed in the following way:

(5.1) The first flash getter used was the barium KIC getter. This consists of barium metal enclosed in an iron tube the wall of which is thin enough to allow a diffusion of barium through the iron at the flashing temperature of the getter. Before flashing, the getters were outgassed at a dull red. The getters were located and flashed in 3 different ways.

(5.11) Two getters were located on either side of the pinch supporting the valve assembly. During flashing of the getters the pressure was kept as low as possible in order to avoid a deterioration of the getter by residual gas ( $p_{\text{man}} < 5 \times 10^{-6} \text{ mm}$ ). After flashing, the getter deposit covered a total area of about  $10 \text{ cm}^2$  on the wall of the glass bulb. It was separated from the main valve assembly by a mica disk possessing 4 holes of  $3.5 \text{ mm}$  diameter. This manner of locating and flashing the getter will be denoted by Ba I.

(5.12) Two getters were located directly beside the valve assembly as shown in Fig. 3a. They were flashed on to the glass bulb in the same way as described in Section 5.11. This getter is denoted by Ba II.

(5.13) Two getters were located as described in Section 5.12, but flashed in an argon atmosphere of about  $5 \text{ mm}$  pressure. In this way, as shown by Ehrke and Slack,<sup>(2)</sup> a black barium deposit is obtained which, owing to its larger surface adsorbs larger quantities of gas than the bright deposit obtained by flashing in a high vacuum. The getter area in this case was  $5 \text{ cm}^2$ . The getter will be denoted by Ba III.

(5.2) The second flash getter used was magnesium. It was made by painting a few mg of magnesium powder together with an organic binder on a nickel tape. This getter was located in the same way as the barium getter in Section 5.12 (Ba II). The magnesium could then be out-gassed and flashed by passing current through the tape. After flashing the getter deposit covered an area of  $8 \text{ cm}^2$ .

(5.3) The first type of coating getter consisted of thorium powder of a particle size between  $1$  and  $10 \mu$ . The powder was deposited, for instance, on the anodes of the valve, by an electrophoretic process which has recently been described by Espe.<sup>(6)</sup> The weight of the deposited coating was  $2.5 \text{ mg/cm}^2$ . After depositing, the coated parts were sintered in a high vacuum at  $1200^\circ \text{K}$  for a few minutes. The thorium getter was located in the valves in two different ways.

(5.31) Location on the two anodes of the valves shown in Fig. 3a (normally on both sides of the anodes). This manner of locating the getter will be denoted by Th I. The total coated area was  $7.5 \text{ cm}^2$ .

(5.32) Location on specially arranged, indirectly heated sleeves as shown in Fig. 3b (denoted by Th II). The coated area in this case was about  $4 \text{ cm}^2$ .

(5.4) The second coating getter consisted of zirconium powder of a particle size between  $3$  and  $20 \mu$ . This powder was deposited by spraying.<sup>(6)</sup> It was sintered and located in the valves in the same way as the thorium. The quantity deposited was again  $2.5 \text{ mg/cm}^2$ .

The experimental valves were supplied with the getters described above and, after sealing to the manifold of the pump, were processed in the following way. A bake of  $60 \text{ min}$  at  $400^\circ \text{C}$  was followed by the decomposition of the oxide cathode and by the usual eddy current heating of the anodes. Subsequently, after a second bake at  $250^\circ \text{C}$  for  $30 \text{ min}$ , the anodes and grids were bombarded with electrons for about  $12 \text{ min}$  at a temperature of  $1100^\circ \text{K}$ . If the coating getters were located on special sleeves (Fig. 3b), these sleeves were heated to  $1100^\circ \text{K}$  simultaneously. The flash getters, if existing, were outgassed, before the second bake was started and were flashed after the electron bombardment.

After this treatment the valves were ready for the measurement of the pressure  $p_g$  above the getters as a function of the manifold pressure  $p_{\text{man}}$ . Normally the manifold pressure was varied to increasing pressures starting from an initial pressure of  $10^{-7} \text{ mm}$ . In all the investigations described here air was used as the gas to be adsorbed by the getter.



## 6. RESULTS

The main results obtained with the different getters are shown in Fig. 6 in which the vacuum factors  $K$ ,

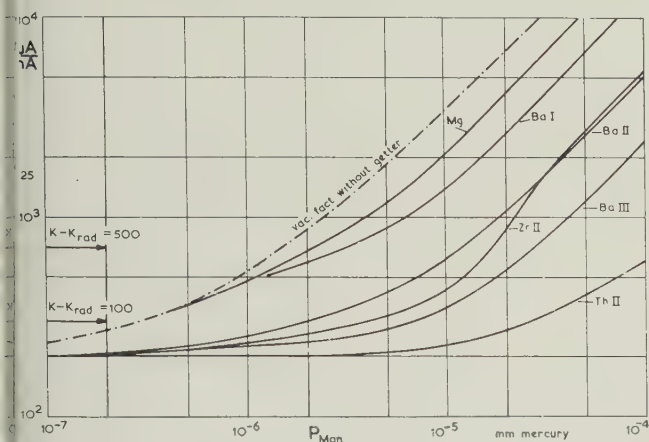


Fig. 6. Vacuum factor  $K$  as a function of manifold pressure for different getters

Mg located at centre of valve	Ba III black deposit located at centre
Ba I bright deposit, located below mica disk	Zr II located on special carriers, $T = 1000^\circ \text{K}$
Ba II bright deposit located at centre	Th II located on special carriers, $T = 1000^\circ \text{K}$

measured in the valves with the getters, are plotted as a function of the manifold pressure  $p_{man}$ . As the radiation level  $K_{rad}$  varies slightly from valve to valve, the curves have been converted to an average radiation level of  $100 \mu\text{A}/\text{mA}$ .

The vacuum factor obtained without any getter, when there is no drop in pressure along the pumping stem, is also plotted in the figure as a dashed curve. For the points on this curve therefore the pressure in the valve is equal to the manifold pressure. Hence the gettering rate of a particular getter is the higher, the more distant the curve, representing this getter in the plot, is from the dashed curve.

If two different getters are to be compared the ratio between the 2 gettering rates  $G_1$  and  $G_2$  can be easily obtained by considering a parallel  $K = \text{const.}$  to the abscissa of Fig. 6. If the manifold pressures, referring to  $K = \text{const.}$  on the two appropriate gettering curves, are denoted by  $p_{man1}$  and  $p_{man2}$ , it follows from equations (3) and (5)

$$\frac{G_1}{G_2} = \frac{p_{man1}}{p_{man2}} \quad (7)$$

This formula allows a comparison of the rates of gettering without any knowledge of the conductance  $F$  of the pumping stem. For a given vacuum factor  $K$  or valve pressure  $p_g$  the ratio between the rates of gettering is equal to the ratio of the appropriate manifold pressures. Considering, for instance, a vacuum factor  $K = 500$ , the rate of gettering of thorium is four times

higher than that of black barium (Ba III) and ten times higher than that of bright barium (Ba II), provided the barium is deposited directly beside the valve system. If the barium deposit is separated from the system by a mica disk, as it is normally done in commercial manufacture, the gettering rate of the barium is only 1/60 of that of thorium. One sees from this that there are considerable differences between the rates of different getters which will certainly influence their practical application. These differences cannot be found if the efficiency of the getters is only measured by determining the adsorbed quantities at rather high pressures. No difference would then, for instance, be found between the two types of barium getters Ba I and Ba II.

A quantitative evaluation of the curves in Fig. 6 has been undertaken for two different vacuum factors, namely for  $K - K_{rad} = 100$  and for  $K - K_{rad} = 500$  (as marked on the ordinate of Fig. 6). These two values of  $K$  correspond to pressures above the getters of  $p_{g1} = 3 \times 10^{-7} \text{ mm}$  and  $p_{g2} = 1.5 \times 10^{-6} \text{ mm}$ . The result has been compiled in the following table (for  $F = 18.5$ ).

Table 1. Type of getter

	Rate of gettering in $\text{cm}^3 \text{sec}^{-1}$	
	$p_g = 3 \times 10^{-7}$	$p_g = 1.5 \times 10^{-6}$
Mg	—	9
Ba I (bright below mica disk)	—	22
Ba II (bright at centre)	125	135
Ba III (black at centre)	400	260
Th II	1 650	1 500
Zr II	230	190

The values given for the thorium and zirconium getters have been measured in valves of the type shown in Fig. 3b, where the metal powder was deposited on special heatable carriers. The temperature of these carriers during the measurement was  $1000^\circ \text{K}$ . The dependence of the rate of gettering on the temperature of the getter will be seen from Fig. 7 in which curves  $K$

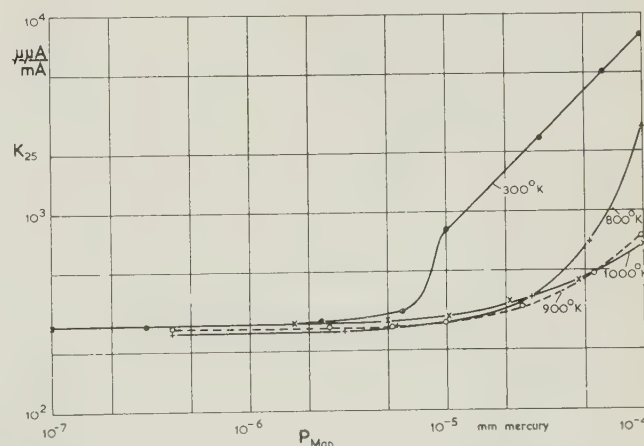


Fig. 7. Vacuum factor  $K$  as a function of manifold pressure  $p_{man}$  for a thorium getter at different temperatures.

against  $p_{man}$  have been plotted for a thorium getter at four different temperatures. At room temperature the high value of the rate of gettering can only be maintained if the manifold pressure, and therefore the quantity of gas to be taken up, are sufficiently low. It is thought that this phenomenon is due to the fact that only an adsorption of gas can take place at room temperature and that the capacity of the getter is therefore easily exhausted. At higher temperatures, however, the gas can diffuse to the interior of the thorium crystals and can be absorbed there. The capacity for gas will therefore be considerably higher, and the high rate of gettering can be maintained much longer. This view is supported by other observations which showed that the gas adsorbed at room temperature could be removed from the getter in a very short time by heating the getter to a high temperature ( $\sim 1\,150^\circ\text{K}$ ). During such a heating a short outburst of gas from the getter was indicated by the ionization gauge. No such outburst could be seen if, after taking up gas at 900 or  $1\,000^\circ\text{K}$ , the getter temperature was raised further to  $1\,150^\circ$ .

The discussion above referred to the thorium getter located on special carriers as shown in Fig. 3b. The results which were obtained with these carriers heated to  $900\text{--}1\,000^\circ\text{K}$  were nearly identical with those for the thorium deposited on the anodes (Th I, see Fig. 3a). This correspondence can be explained by the fact that the surface temperature of the thorium crystals in the coating will be much higher than the temperature of the metal carrier underneath, due to the loose agglomeration of the thorium particles in the coating. During the bombardment of the anodes to a dull red it could, for instance, be seen that the thorium crystals which were directly hit by the bombarding electrons had a considerably higher temperature (bright red) than those which were in the shadow of the screen grid wires. It can therefore be assumed that in the case of thorium deposited on the inside of the anode the surface temperature is sufficiently raised to allow diffusion and absorption to take place.

The curves obtained for heated thorium and for thorium deposited on anodes were well reproducible. Table 2, giving  $K - K_{rad}$  for three different valves with thorium on both sides of the anodes and for another valve with the thorium on a special carrier shows this

Table 2. *Values of  $K - K_{rad}$  for thorium getters arranged in different ways*

$p_{man}$	Thorium on both sides of anode				Thorium on special carrier	Thorium on inside of anode	Thorium on outside of anode
	I	II	III	Average	$1\,000^\circ\text{K}$		
$10^{-6}$	0	0	0	0	0	2	50
$3 \times 10^{-6}$	12	7	10	10	5	10	110
$6 \times 10^{-6}$	32	20	23	25	12	25	430
$10^{-5}$	57	40	45	47	28	50	1 150
$3 \times 10^{-5}$	173	145	165	161	115	155	5 400
$6 \times 10^{-5}$	338	300	335	324	240	285	$\sim 10\,000$
$10^{-4}$	538	485	535	519	400	435	—

reproducibility. The reproducibility during several measurements of a particular valve was as good. The table also gives values of a valve in which the thorium was only deposited on the inside of the anode, and for another one in which only the anode outside was coated. It will be seen that a thorium coating only on the inside is as effective as a coating on both sides, whilst a coating only on the outside is very ineffective. This will be partly due to the fact that the coating on the outside is not heated by the bombarding electrons. Another responsible factor is the wider distance between the outside of the anode and the regions where the gas molecules are exposed to the impact of electrons. This distance will influence the efficiency of the thorium getter in the same manner as that of the barium getter is influenced.

Finally we may look back to Fig. 1 which shows the vacuum factors measured in valves with different getters after sealing-off. In general the results plotted in Fig. 1 agree fairly well with the values of the gettering rate which have been measured (see Table 1). The thorium getter which has the highest gettering rate gives the lowest initial pressure in the valves, barium (Ba II) with a medium rate gives medium pressures and magnesium with a very low rate gives high pressures. The only getter which shows slightly contradictory results is zirconium. Considering its gettering rate, which is in the medium range, the valves made with a zirconium getter should have lower initial pressures than those shown in Fig. 1. This different behaviour has not yet been explained.

A curve in Fig. 1 which has not yet been discussed is that referring to a solid thorium anode. The pressures in a valve made with such an anode (thorium sheet  $0.005$  in thick) are much higher than those measured with a thorium getter. A high gettering rate can only be obtained if the thorium is used as a powder which shows that the physical structure of this powder is important for its gettering efficiency.

## 7. CONCLUSIONS

The above investigations show that the efficiency of getters at low pressures, as measured by their rate of gettering, varies widely between different getters. A variation by a factor of more than 100 has, for instance, been found between the rates of magnesium and thorium getters. Furthermore, the gettering rate depends on the location of the getter in the valve and, for instance, in the case of barium, can be increased by a factor 6 by moving the getter from its normal position outside the assembly cage as near as possible to the centre of the assembly. This increase in efficiency may show that gas ions or atoms are more readily taken up by a getter than molecules. If the getter is more distant from the region of the valve where the ions or atoms are produced a part of these will recombine to molecules and the efficiency of the getter will therefore fall. It has always



been assumed that such an influence exists, but it has not been possible to measure it and to give figures for its magnitude.

It is interesting to compare the values, measured for the rate of gettering, with the pumping speed one can obtain by using a diffusion pump. At pressures of about  $10^{-6}$  mm or lower the fall in pressure along the pumping stem of the valve will be very small. Assuming  $p_g = 2 p_{man}$  and a pumping stem of the same dimensions as in our experiments (conductance  $F \simeq 20 \text{ cm}^3 \text{ sec}^{-1}$ ), the pumping speed would be  $40 \text{ cm}^3 \text{ sec}^{-1}$ . This only holds on the supposition that the speed of the diffusion pump is large compared with the conductance of the pumping stem. The speed of diffusion pumps is well below  $1\,000 \text{ cm}^3 \text{ sec}^{-1}$  at a pressure of  $10^{-6}$  mm<sup>(7)</sup> and decreases rapidly with decreasing pressure. At pressures lower than  $10^{-6}$  mm, the actual pumping speed with which the valve is evacuated will therefore be even lower than the above value of  $40 \text{ cm}^3 \text{ sec}^{-1}$ . When comparing this with the gettering rates of more than  $1\,000 \text{ cm}^3 \text{ sec}^{-1}$  which are attainable at these pressures, one recognizes the importance of getters for achieving such low pressures within a reasonable time.

Further investigations have now been started in which the influence of the location of the getter, the influence of ionization and the decay of a getter during its operation will be examined in more detail. These experiments are being extended to all the gases which can be expected to compose the residual gas in a valve.

A factor bringing about a certain inaccuracy in the

present measurements, especially at very low pressures, is the existence of the radiation level in the normal ionization gauges. A special ionization gauge in which the radiation level is reduced to zero has recently been designed by Dr. G. H. Metson (1950b) of these laboratories, and this gauge will therefore be used in the future experiments.

#### 8. ACKNOWLEDGEMENT

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# Characteristics of Diffusion Pumps

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The essential characteristics of a diffusion pump are discussed, and apparatus and methods for determining them are described. The comparison of the performance of different pumps is discussed, and as an example experimental results are given for the relation between performance and gap width. Finally, theories of diffusion pump operation are discussed in the light of these experimental results and some future lines of experiment indicated.

## INTRODUCTION

In a recent paper<sup>(1)</sup> it was shown that the action of a pump could be better appreciated by the introduction of the terms "intrinsic speed" and "operational speed." In this paper it is proposed to show how the significance of these terms can be demonstrated experimentally; these ideas will then be applied to a discussion of the most important characteristics of a diffusion pump and a description given of the most convenient methods for determining them experimentally.

## CHARACTERISTICS TO BE DETERMINED

It is first necessary to emphasize that the intrinsic speed is an attribute of the pump itself, whilst the operational speed is a function of the system, since it may depend on the particular backing pump used. Although a knowledge of the operational speed is extremely useful in many practical cases, and details of how to determine it will be given later, yet it must be realized that the inherent properties of a pump, and a comparison of the properties of different pumps, can only be obtained by means of a determination of the intrinsic speed.

What, then, are the most important characteristics of a diffusion pump? For a given pump, with a given operating fluid, the variables are intrinsic speed, fine side pressure, critical backing pressure and heat input. Of these the last mentioned has probably the least fundamental significance, and hence a most convenient family of characteristics is that of intrinsic speed against critical backing pressure for a series of fine side pressures. Probably the most convenient way to obtain this data experimentally is to determine, for a number of heater wattages, the relation between fine side pressure and backing pressure for a number of discrete values of throughput (speed  $\times$  pressure). From these curves it is possible to deduce various characteristics. In some cases, however, it is advisable to obtain a desired characteristic directly, simply for reasons of experimental expediency. This was necessary when experiments were carried out to show the meaning of intrinsic and operational speeds, and for this reason they will be described separately.

## DESCRIPTION OF APPARATUS

The apparatus (Fig. 1) consisted of an approximately

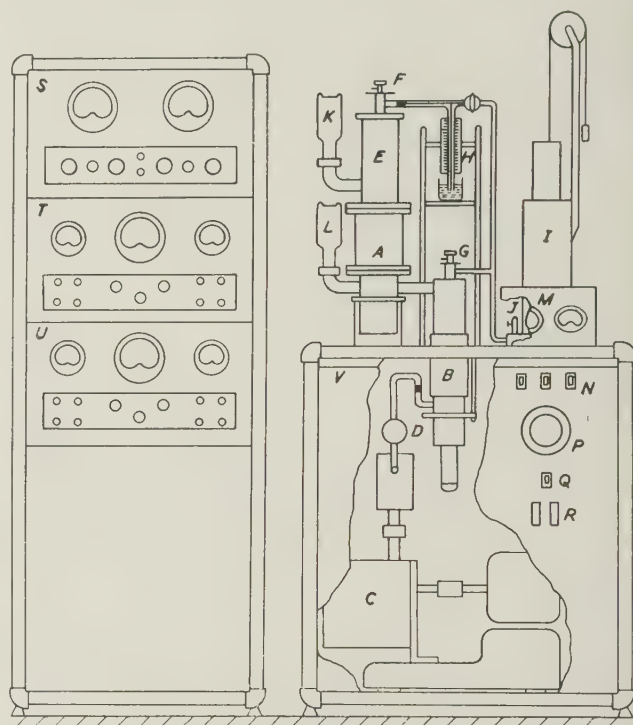


Fig. 1. Apparatus for finding characteristics of diffusion pumps

- |   |                             |   |                              |
|---|-----------------------------|---|------------------------------|
| A | Experimental pump.          | L | Bkg. side ionization gauge.  |
| B | O3B diffusion pump.         | M | Voltmeter and ammeter.       |
| C | D.R.I. rotary pump.         | N | Pump switches.               |
| D | Vacuum valve.               | P | "Variac."                    |
| E | Mixing chamber.             | Q | Main switch.                 |
| F | Fine side needle valve.     | R | Fuses.                       |
| G | Bkg. side needle valve.     | S | Pirani gauge controls.       |
| H | Tetralin flowmeter.         | T | Fine side ion gauge control. |
| I | Gas reservoir.              | U | Bkg. side ion gauge control. |
| J | To nitrogen cylinder.       | V | Shelf for note-book.         |
| K | Fine side ionization gauge. |   |                              |

3-in diameter single stage oil diffusion pump, fitted with an immersion heater and backed by a Metrovac O3B oil diffusion pump which in turn was backed by a Metrovac DR1 rotary oil pump. A standard Metrovac O3 jet system<sup>(2)</sup> was used in the experimental pump. A number of cylindrical inserts were made: these all had the same outside diameter so as to slide inside the pump, but had different wall thicknesses, so that in effect the gap width of the pump could be varied without alteration to the jet system.



Nitrogen could be admitted to the pump through a flowmeter and a needle valve at the top of a small chamber fitted above the diffusion pump, which, incidentally, was not fitted with baffles. The flowmeter\* consisted of a capillary tube dipping into a beaker of tetralin, and the throughput was measured by timing the rise of tetralin from one mark to another. The fine side pressure was measured by means of a Mazda 29D2 ionization gauge attached to a side tube which entered the mixing chamber at the bottom, i.e. just above the jet system of the pump. As in this series of experiments the emphasis was on relative measurements, a standard gauge factor was assumed; for accurate absolute values each gauge would require to be calibrated. Backing pressures were measured by means of either a second ionization gauge or a water-cooled Pirani gauge situated

normal backing duct. In this way the gauge indicated directly the backing pressure without the necessity for the application of a correction factor. A second needle valve on the backing side of the pump enabled varying amounts of nitrogen to be admitted so that the backing pressure could be kept constant.

Experience has shown that this equipment, which is suitable for research or routine measurements on pumps or for educational purposes, is best built up on the unit principle. On one framework, or cubicle, approximately  $3 \times 2 \times 2\frac{1}{2}$  ft high, could be mounted the experimental pump, the backing pump and the rotary pump, together with the associated gauges and flowmeter (see Fig. 1). On it could also be conveniently placed the switches for the pumps, and the Variac, voltmeter and ammeter for controlling and measuring the heater wattage applied to the experimental pump. A second cubicle, approximately  $2 \times 1 \times 4\frac{1}{2}$  ft high, suitable for the mounting of standard 19-in chassis, could house the Pirani gauge control gear and the two sets of control gear for the two ionization gauges. If measurements at high fine side pressures are contemplated; then it might be advantageous to fit two sets of Pirani gauge control gear side by side on a single 19-in chassis. With this general arrangement of apparatus it is possible for a seated operator to take all the measurements.

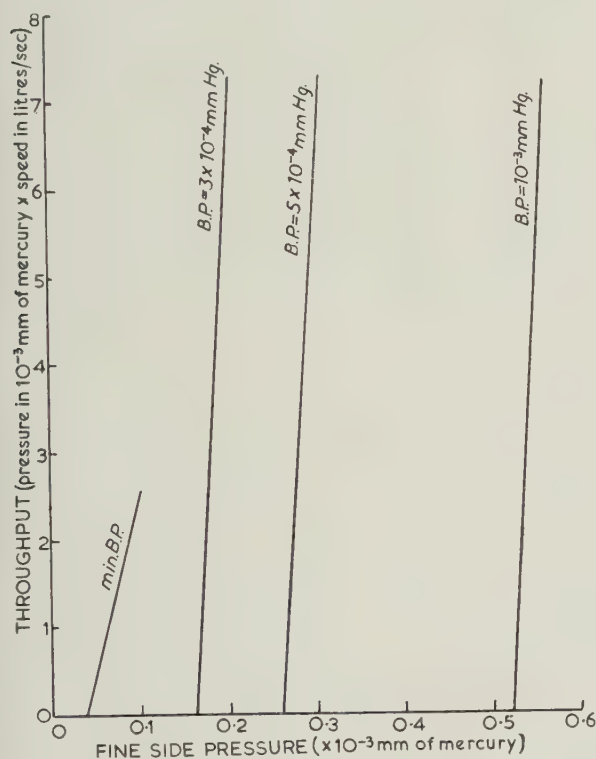


Fig. 2. Diffusion pump characteristics.  
Heat input 51 W.

immediately above the backing pump. Corrections were applied for the pressure difference between the ends of the pipe connecting the experimental pump to the backing pump. In some of the later experiments this pipe was replaced by a rectangular duct of much larger cross-sectional area, and at the same time the gauge was attached to a pipe which entered the pump opposite the

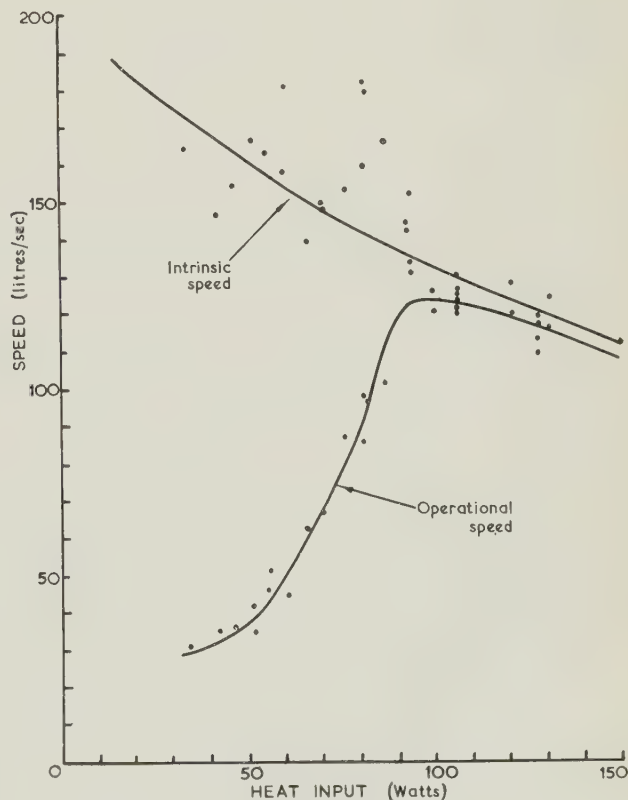


Fig. 3. Variation of speeds with heat input

\* Of the type described by C. W. Miller in his M.Sc. thesis entitled "Some Experiments with the Oil Vapour Diffusion Pump," University of Leeds, April 1939.

## EXPERIMENTAL METHODS AND RESULTS

*Significance of Intrinsic and Operational Speeds.*—Measurements were made, for a given heater wattage, of throughput against fine side pressure, for a series of constant backing pressures obtained by adjustment of the needle valve on the backing side of the pump. A further characteristic was obtained when the needle valve on the backing side was kept closed, thus simulating ordinary practical conditions. A typical set of results for different backing pressures (B.P.) is shown in Fig. 2, from which it can be seen that whilst the operational speed (i.e. the slope of the min. B.P. line)\* is 42 l/sec, the intrinsic speed (i.e. the slope of the constant B.P. lines)† is about 167 l/sec. When the backing

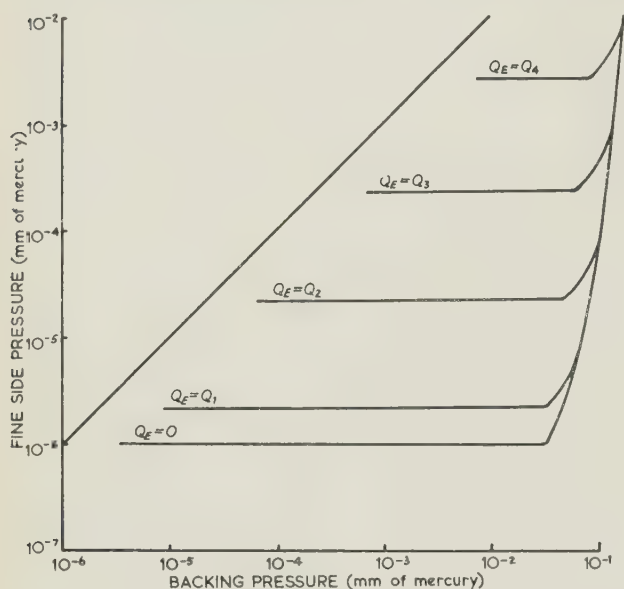


Fig. 4. Variation of fine side pressure with backing pressure. Gap width 1.5 mm. Heat input 134 W.  $Q_1$ ,  $Q_2$ , etc., refer to various discrete values of the external throughput  $Q_E$ .

side needle valve was closed, the backing pressure rose as the throughput was increased. If the pump were being operated under such conditions that the fine side pressure were sensibly independent of the backing pressure, then the slope of the throughput/fine side pressure characteristic would be the intrinsic speed. It can be seen from Fig. 2, however, that as the backing pressure rose as the result of increasing throughput the partial fine side pressure due to back-diffusing gas also rose, resulting in a line whose slope was less than that corresponding to the intrinsic speed. The smaller slope of this curve, i.e. the smaller value of the operational speed, was therefore due to the pump being operated under such conditions that the critical backing pressure was being exceeded.

\* From equation (13) and the following paragraph of the earlier paper.<sup>(1)</sup>

† From equation (9) of the earlier paper.<sup>(1)</sup>

Similar sets of measurements were made at other heater wattages, and Fig. 3 shows the relation between intrinsic and operational speeds and heater wattage. At low heater wattages it was difficult to obtain stable equilibrium conditions; individual intrinsic speed determinations were therefore not very reproducible, but it is clear from Fig. 3 that there is a definite tendency for the intrinsic speed to rise continuously as the heater wattage is reduced, whilst the operational speed passes through a maximum value. In order to obtain the fundamental characteristics of the pump, and not those of the system, it is therefore necessary to work either at constant backing pressure or below the critical backing pressure.

*Determination of Principal Characteristic Curves.*—As indicated earlier, the experimental approach was to obtain at various heater wattages, fine side pressure-backing pressure characteristics at different fixed values of throughput. Two such series of curves are shown in Figs. 4 and 5. From Fig. 4 it can be seen that there is a range of conditions where the backing pressure is below the critical value, whereas Fig. 5 shows that, under these particular conditions, the critical backing pressure is always exceeded. Although the intrinsic speed can be obtained from either series, it was considered desirable in this case as well to make separate determinations of throughput against fine side pressure

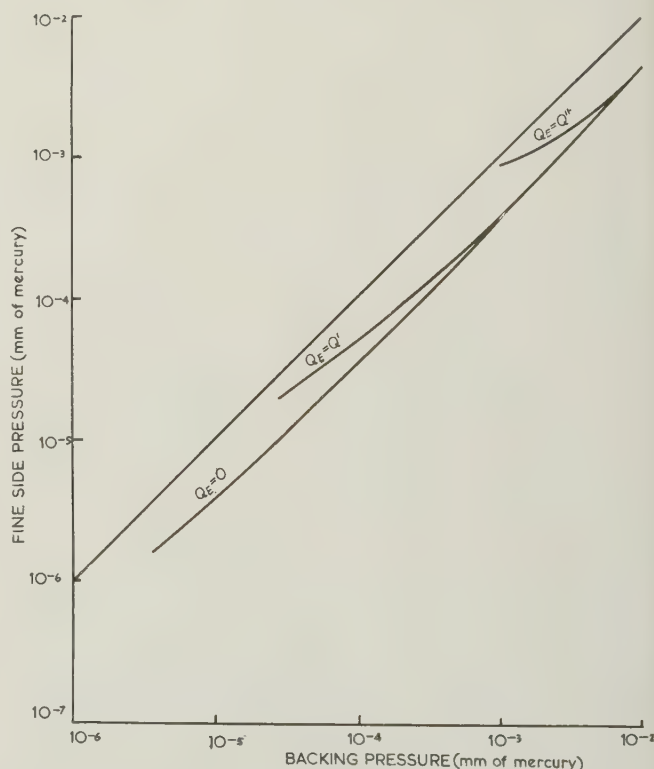


Fig. 5. Variation of fine side pressure with backing pressure. Gap width 1.5 mm. Heat input 38 W.  $Q'$ ,  $Q''$ , etc., refer to discrete values of the external throughput  $Q_E$ .



so that the most accurate values of intrinsic speed could be obtained. As it was rather tedious to work at constant backing pressure, measurements were only made in those cases where the critical backing pressure was not exceeded, as shown by the earlier measurements. Such values of intrinsic speed are very useful, moreover, as they can be obtained under normal working conditions; in fact, under these conditions, the intrinsic speed is the same as the operational speed. Values obtained under conditions such as those shown in Fig. 5, on the other hand, are of academic interest only.

Hence the relation between intrinsic speed and heater wattage could be obtained for a particular value of fine side pressure. As the relation between critical backing pressure and heater wattage could also be determined, it was possible to deduce the curve showing the relation between intrinsic speed and critical backing pressure for the particular fine side pressure. This curve, marked "1.5 mm gap," is shown in Fig. 6, which will be discussed in detail later.

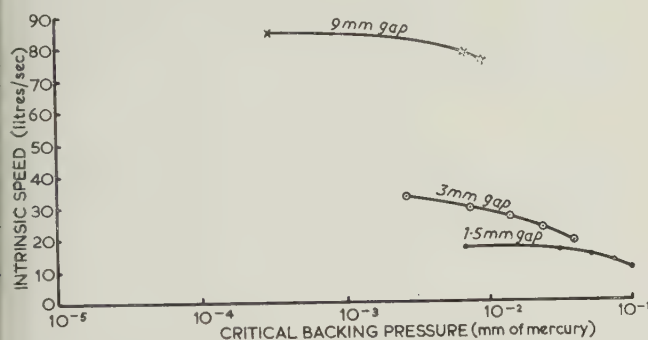


Fig. 6. Variation of intrinsic speed with critical backing pressure. Fine side pressure  $5 \times 10^{-5}$  mm of mercury

*Comparison of Characteristics of Different Pumps.*—The characteristic just mentioned, namely intrinsic speed against critical backing pressure, is probably the most suitable to use when comparing the performances of different pumps, such as those with different gap widths, different designs of jet, different pump fluids, etc. As an example, a comparison of the performances of a pump with a given jet system and operating fluid but with various gap widths will be given here. The constructional details of the system have already been given in the section "Description of Apparatus." The results are shown in Fig. 6, which gives the data relating to three values of gap width. It may be interesting also to give the curves for efficiency against critical backing pressure (Fig. 7) and efficiency against heat input (Fig. 8). Efficiency is defined as the ratio of the intrinsic speed to the speed of a perfect pump, i.e. the conductance of the annular gap.

#### DISCUSSION

Since intrinsic speed and efficiency depend on the heat input it follows that it is not possible to specify a pump

by any single value. Care must therefore be taken in assessing the relative efficiencies of pumps for which only single values have been quoted in the literature. The apparent superiority of one jet system over another, for example, may in reality be due more to the operating conditions than to the jet system itself; similar remarks apply to the comparison of the performance of pumps with different fluids.

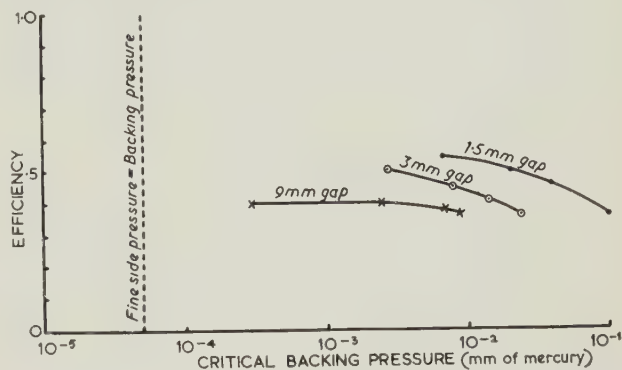


Fig. 7. Variation of efficiency with critical backing pressure. Fine side pressure  $5 \times 10^{-5}$  mm of mercury

There has recently been a great interest taken in the speed of a diffusion pump for hydrogen. It has been shown<sup>(3), (4)</sup> that anomalous results could be explained by the fact that hydrogen tends to back diffuse through a pump more readily than nitrogen. If back diffusion occurs to a large extent, it is due to the backing pressure being above the critical value, and, as has already been

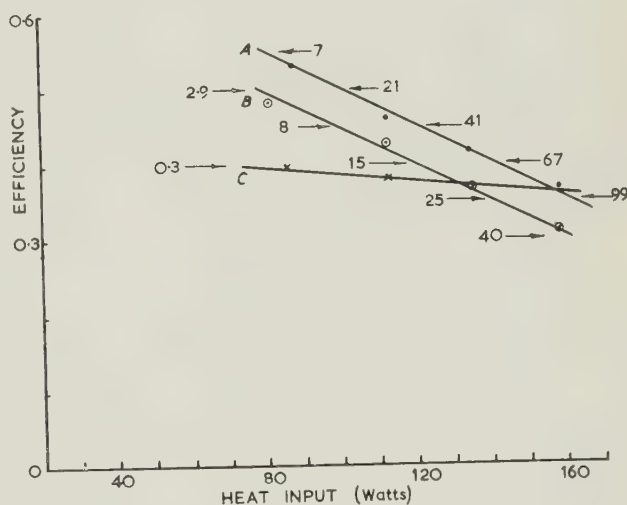


Fig. 8. Variation of efficiency with heat input. Fine side pressure  $5 \times 10^{-5}$  mm of mercury. Gap widths:

Curve A 1.5 mm.  
Curve B 3.0 mm.  
Curve C 9.0 mm.

Figures on curves indicate backing pressures in  $10^{-3}$  mm of mercury.

pointed out, measurements taken under these conditions are representative of the system rather than the pump. Blears and Hill<sup>(4)</sup> sum up by saying that the inverse root law for gases of different molecular weight is obtained "provided the pump is operated in such a way that no molecules can return in a reverse direction through the jet." It is now suggested that this statement can be generalized by saying that the ratio of the intrinsic speeds for different gases is given by the inverse ratio of the square root of the molecular weights of the gases. The ratio of the operational speeds may be any lower value depending entirely on the particular conditions obtaining in the test.

It may be interesting to discuss now Gaede's statement<sup>(5)</sup> that the optimum speed of his diffusion pump was obtained when the mean free path  $\lambda$  of the gas molecules in the backstreaming vapour molecules was equal to the gap width  $d$ , i.e. when  $\lambda/d = 1$ . This gives the impression that fundamentally, for a given  $d$ , there is a critical value of  $\lambda$  and thus a critical value of heat input, for optimum speed, or, similarly, for a given  $\lambda$  and thus heat input, there is a critical value of  $d$ .

It must be realized first that, as shown recently,<sup>(1)</sup> the speed defined by Gaede was neither operational speed nor intrinsic speed, but rate of exhaustion. Like operational speed, rate of exhaustion depends on the characteristics of the pumping system as a whole, so that any conclusions reached by Gaede regarding rate of exhaustion were dependent, amongst other things, on the particular rotary pump he used to back his diffusion pump. The use of rate of exhaustion cannot therefore lead to any conclusions regarding the fundamental behaviour of the diffusion pump itself, but only to the results obtained in specific cases. Let us therefore consider the matter afresh from the point of view of intrinsic speed, i.e. the speed of the pump under conditions when the backing pressure has no appreciable effect.

Fundamentally, Gaede<sup>(5)</sup> showed that the volume of gas passing per second from the fine side to the backing side, or intrinsic speed, is given by

$$S_I = k\alpha A/\sqrt{2\pi\rho_0} \quad (1)$$

where  $A$  is the area of the diffusion gap,  $\rho_0$  is the density of the gas at the working temperature under unit pressure,  $k$  is the absorptivity factor, and  $\alpha$  is a function of  $\lambda/d$  which increases as  $\lambda/d$  increases. The form of the relation between  $\alpha$  and  $\lambda/d$  is given in Fig. 9. For a perfect pump

$$S_0 = A/\sqrt{2\pi\rho_0} \quad (2)$$

so that the efficiency  $\eta_g$  is given by

$$\eta_g = S_I/S_0 = k\alpha \quad (3)$$

The efficiency of the pump therefore depends directly on the value of  $\alpha$ . For a pump with a given gap width  $d$ ,  $\alpha$  tends asymptotically to unity as  $\lambda$  is increased, i.e. as the heat input is reduced. Thus the efficiency of the

pump steadily increases as the heat input is reduced, there being no maximum value when  $\lambda = d$ , although it can be seen that at this point  $\alpha$  has very nearly reached its asymptotic value. Similarly, if the heat input is kept constant, but the gap width is decreased, there is also a steady increase in efficiency without there being any maximum value, although there may be a value which the efficiency approaches asymptotically.

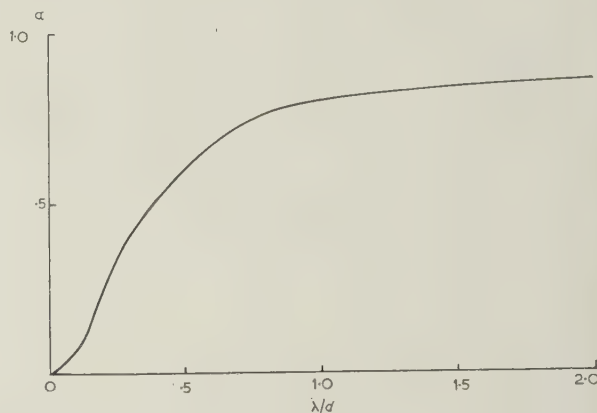


Fig. 9. Relation between mean free path  $\lambda$  and gap width  $d$ , equation (1)

The first of these conclusions is in accordance, at least qualitatively, with the results shown in Fig. 3, where it can be seen that the intrinsic speed, which for a given gap width is proportional to the efficiency, continuously increases as the heat input is reduced. On the other hand, as the heat input is reduced, the operational speed begins to fall at a certain heat input, as for small values the critical backing pressure is exceeded; the critical heat input, however, is only dependent on the particular backing pump used, and the value has no fundamental significance.

Some support for the second conclusion is given by the curves shown in Fig. 8; for at least low heater wattages, for any fixed value, the efficiency increases as the gap width is reduced. Similar determinations of the variation of speed factor or efficiency with gap width have previously been made by Copley and others,<sup>(6)</sup> but it is rather difficult to draw any simple conclusions from their experiments, as the boiler pressure head was not kept constant for the various gap widths.

At low heater wattages, when the vapour velocities are small, and the vapour is more or less "diffusing" out of the jet, the efficiency depends on the gap width, as already mentioned. In these circumstances it might be conjectured that the efficiency does not depend to any great extent on the shape of the jet. The results of some preliminary experiments carried out some years ago by D. G. Avery and the author tend to support this view. In these experiments the divergence of the vapour nozzle was changed without any alteration being made to the gap width or path followed by the incoming gas molecules. It was found that there was no significant



change in the efficiency of the pump. Obviously, however, this point needs more investigation.

At high values of heat input, on the other hand, when the vapour stream emerges with more energy, the reverse may be true, i.e. the efficiency may depend more on the shape of the jet and less on the gap width. The former suggestion is borne out by the fact that in ejector pumps, where the heat input is great, the shape of the jet is very important. Although no definite conclusions can be drawn from Fig. 8, as an insufficient number of experiments have been made, yet it does give some support for the view that at high heater wattages the efficiency is less dependent on the gap width. More convincing evidence on this point has, however, been given by Alexander,<sup>(7)</sup> who showed that, although the efficiency decreased as the heat input was increased, yet, at high heater wattages, the efficiency was independent of the gap width.

There thus appears to be the possibility that at low values of heat input the diffusion principle is important and that as the heat input is increased a gradual transition occurs until at high values of heat input the ejector principle determines the action of the pump. In this connexion it may be noted that Alexander said that his work directly contradicted Gaede's diffusion principle. In a postscript to a paper on the theory of the diffusion pump, Jaeckel<sup>(8)</sup> disputed this statement and argued that Alexander's results obtained at high pressures would be in accordance with his own theory at high pressures (not yet published), which was based on Gaede's conceptions. If, as conjectured above, there is a transition from Gaede's diffusion principle to the ejector principle as the heat input is increased, difficulties may disappear if practical results are compared with the relevant theory. In this case, Gaede's theory and Alexander's theory may well be complementary since they apply to different regions of operation. Further

work, both theoretical and experimental, is required to enable a complete solution to be obtained.

## CONCLUSION

In the past, the specifications of the performance of many pumps have been given in the literature, but there does not appear to have been any common basis. As progress depends on true evaluation and comparison, it is essential that a universally applied method of specification is used, and it is hoped that the methods described in this paper may help somewhat in the search for the ideal method.

## ACKNOWLEDGEMENTS

I should like to acknowledge the help given me during the progress of this work by many of my colleagues, particularly Messrs. D. G. Avery and C. A. Taylor, who carried out the greater part of the experimental work.

I should also like to express my thanks to Sir Arthur P. M. Fleming, Director of Research and Education, and to Mr. B. G. Churcher, Manager of Research Department, Metropolitan-Vickers Electrical Co. Ltd., for permission to publish this paper.

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# A Note on the Improvement of the Field of a Solenoid by End Coils

By B. LEWIS, B.Sc., University of Nottingham

[Paper received 1 May, 1950]

The addition of end coils to a solenoid considerably improves the uniformity of the axial field. Design procedure is given for the general case, and details of the dimensions and performance of a solenoid constructed to a given specification are quoted.

J. R. Barker<sup>(1)</sup> has recently given formulae and tables for computing the uniformity of the field inside a solenoid. When a uniform magnetic field is required over a length which is long compared with the diameter of the solenoid, an appreciable economy in solenoid length may be made possible by the use of two uniformly wound correcting coils carrying the same current as the main solenoid winding. Such a field is often needed to magnetize a long, thin rod specimen. Correction of the field of a solenoid by end coils has been used by several workers,<sup>(2)</sup> but details of design procedure and performance do not seem to be available.

As an example, the axial field in a simple solenoid of length 17 units having inner and outer radii  $a = 1$ ,  $b = 2$  respectively is constant to 0.5% over the central 6 units. If suitable correcting coils are added, the axial field may be made uniform to 0.5% over the central 12 units.

The dimensions of the solenoid and correcting coils needed in any given case can be obtained theoretically by trial. For a chosen length of solenoid, the field distribution along the axis can be calculated, and plotted as in Fig. 1 (full-line), either by Barker's formulae, or by approximating a thick solenoid to the sum of several thin coaxial solenoids, using the simple  $2\pi ni(\cos \theta_B - \cos \theta_A)$  formula. The correcting coils will be thin coils, one at each end of the solenoid, with inner radius  $c$ , slightly larger than the outer radius of the solenoid, and outer radius  $d$ , to be determined. They can be approximated to several elementary concentric coils of radii  $c + \frac{1}{2}\epsilon$ ,  $c + 3/2\epsilon$  . . .  $d - \frac{1}{2}\epsilon$ , where  $\epsilon$  is small compared with  $c$ , and the field at points along the axis of the system calculated for each of these elementary coils. The field distribution along the axis for any value of  $d$  is obtained by summation and is then reversed in sign and plotted, as in Fig. 1 (broken line), such that three points, representing the fields at the middle and near each end of the desired uniform region, are coincident with the corresponding points on the solenoid field distribution curve. The resulting field due to the solenoid and correcting coils is obtained by the addition of the individual fields of each. For convenience in graphical analysis, the field of the correcting coils has been reversed in sign before plotting, and hence the difference between these curves represents the remaining non-uniformity of the resultant field. The number of

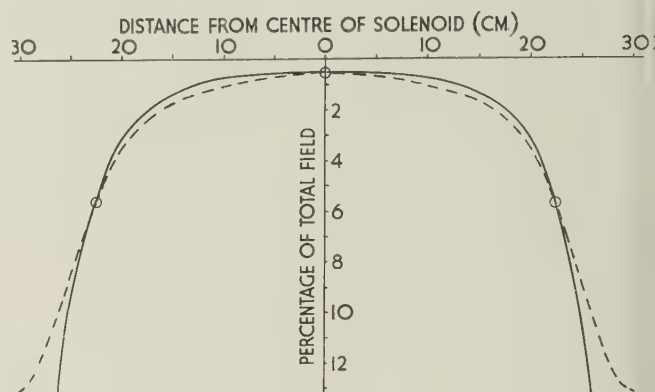


Fig. 1. Calculated field distributions.

———— Simple solenoid.  
----- Correcting coils (field reversed in sign).

turns per layer of the correcting coils is still arbitrary (though it must be small), and is chosen to satisfy the condition that the coils shall carry the same current as the solenoid.

A solenoid of length 60 cm,  $a = 3.5$  cm,  $b = 5.5$  cm, for use with a specimen 40 cm long, has recently been designed and constructed. It was found that an extremely good fit of the two curves was easily obtained, and for  $c = 7$   $d = 11$  cm the agreement of the curves along the axis was better than 0.1% of the total field over two-thirds of the length of the solenoid. The values of  $c$  and  $d$  were not critical. Forty layers each of ten turns were required in the correcting coils compared with twenty layers each of five hundred turns in the main winding. The axial length of the correcting coils was 1 cm. They were initially wound with rather more than the calculated number of turns, and, shunted by variable resistors, were connected in series with the main solenoid winding. Field measurements were then made and the shunts adjusted for optimum uniformity of the field. Finally, turns were removed to give the same field without shunts.

A differential ballistic method was used to test the uniformity of the field, using two identical search coils, connected in series opposition, one being at the middle of the solenoid  $O$ , and the other at any desired point  $X$ . The galvanometer deflexion on making or breaking the field current measured the difference in field between



and  $X$ . To plot the field near the centre the fixed coil was moved to another position as a temporary datum and the readings subsequently obtained corrected by a figure representing the difference between the field at  $O$  and at the temporary datum position. The search coils used had 1 000 turns of mean radius 0.6 cm and breaking a field current of 2 A gave 30 mm deflexion for a field difference of 1%. This calibration figure was obtained by measuring the galvanometer deflexion produced by one coil only when a field current of 20 mA was broken.

Measurements of the axial field of the solenoid described above and of three other solenoids showed distinct asymmetry, and kinks in the curve departing in some cases by as much as 0.5% from the smooth symmetrical curve which was expected. Winding in each case had been by hand-feeding the wire on to the solenoid turning in a lathe and any variation in tension or thickness of insulation would lead to irregular spacing of the winding. The final field distribution for the solenoid of which details have been given is shown, with the uncorrected field, in Fig. 2, and the 0.4% non-uniformity is ascribed principally to winding irregularities. It would appear that if a very uniform field is required, extreme care must be taken during winding and that machine-feeding is essential.

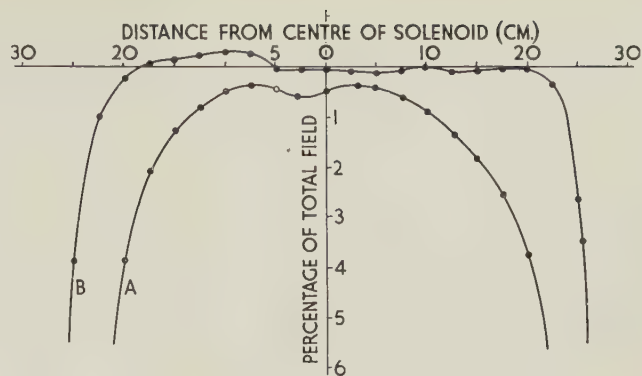


Fig. 2. Measured field distributions.

Curve A—Solenoid without correcting coils.  
Curve B—Same solenoid with correcting coils.

Acknowledgement is made to Professor L. F. Bates for the facilities of the laboratory and to Dr. R. Street for encouragement and advice.

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## NOTES AND NEWS

## Correspondence

**The Pirani Effect in a Thermionic Filament as a Means of Measuring Low Pressures**

Although in the work carried out at Exeter and described in the paper which I communicated to you and which appeared in your May issue, Dr. L. Speirs (not Spiers) was associated with Mr. Jolly, he is not, in fact, a joint author of the paper. As Mr. Jolly failed to consult Dr. Speirs he should, of course, have omitted the name from the heading of the paper.

This, therefore, is to state that Mr. Jolly alone is responsible for the published paper, and any inconvenience caused to Dr. Speirs is regretted.

J. THOMSON.

Royal Naval College, Greenwich.

*We wish to associate ourselves with Professor Thomson's expression of regret and apologize to Dr. Speirs for the inconvenience caused to him by our failure to verify that he had personally approved of the paper before publication. We understand that Dr. Speirs had intended to publish an account of their work jointly with Mr. Jolly; but we are informed that he disagrees with certain statements and opinions expressed in the paper which appeared in our May issue.*

EDITOR.

**"Survey of General and Applied Rheology"**

Dr. Lawrence's review of Scott Blair's *Survey of General and Applied Rheology* which appeared in your June issue

invites comment. The name "rheology" is no more "esoteric" (in the senses defined by Webster and O.E.D.) than a score of other "ologies" in current use. Both derivation and meaning are clear, and the prefix "rheo-" is well established in Webster's Dictionary of 1928.

It is not clear what your reviewer means by "an esoteric outlook." Rheology is not reserved only for the initiated. Scott Blair's book has a psychological and philosophical bias, but its aim and outlook have little in common with schools of occult and mystical (as defined by Webster and O.E.D.) thought. The sentence "Rheology will never become a science until it ceases to try to be rheology" is meaningless as it stands. Presumably Dr. Lawrence objects to any departure from classical physics and the introduction of psychological methods; but it is not unscientific to read psychology and certain problems of undoubtedly industrial importance cannot be solved by purely physical methods.

Your reviewer is entitled to his poor opinion of the book in question; but I hope this will not deter others interested in problems of deformation and flow of matter from examining it for themselves. They may find it less inept and unsatisfactory than they have been led to believe.

V. G. W. HARRISON.

Little Bookham, Surrey.

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## SPECIAL REPORT

## Summarized Proceedings of a Conference on Stress Analysis London, 1950

A Conference of the Stress Analysis Group of The Institute of Physics was held at University College, London, on 3-5 April, 1950. There were five sessions devoted to the following subjects: Development work on a Brittle Lacquer suitable for experimental Stress Analysis, The use of models in Structural work, The application of experimental Stress Analysis methods to Soil Mechanics problems, Recent developments in Photoelasticity, and The application of experimental Stress Analysis methods in the Motor Industry. Papers are summarized in this report.

## BRITTLE LACQUERS

MR. C. H. YOUNG (Paints Division, Imperial Chemical Industries Ltd.) read a paper on "Development Work on a Brittle Lacquer suitable for Experimental Stress Analysis" during the opening session of the Conference. This work had been carried out in response to a demand for a strain sensitive lacquer which would be readily available in this country.

Mr. Young said that the value of using a continuous strain indicating coating for the determination of surface strains in mechanically tested models and prototypes appeared to be generally accepted. This form of strain indication was of greatest value if the required pattern could be obtained without exceeding the elastic limit of the material, so that the tests are non-destructive. In design work where the relative stress concentrations were often more important than the absolute values, a strain indicating coating could give a rapid pictorial survey of the stresses at the surface of the object under test. This information could, if necessary, be supplemented by the use of strain gauges which would then be correctly located for the determination of the absolute value of the strain. Semi-quantitative results could, however, be obtained by use of the coating alone, by reference to a suitable calibration strip which was strained alongside the test object. The lacquer when strained beyond a pre-determined value formed cracks at right angles to the direction of the principal tensile strain. Compression stresses could, however, also be determined by using relaxation loading methods, but it had not so far been possible to examine Strainlac (which was the name given to the new lacquer) for this purpose.

Many attempts to prepare a brittle lacquer had been reported in the literature, by the Germans as early as 1932 and later by the Russians; but the most successful range which it is stated has given excellent quantitative results was marketed in America by the Magnaflux Corporation under the name "Stresscoat." British Patent 565210, accepted in 1944, covered such lacquers, and specified a very wide range of film-forming materials, natural resins, processed natural resins and synthetic resins. The particular plasticizers mentioned were dibutyl phthalate and *n* butyl stearate and the solvents

carbon disulphide, methylene dichloride and 1,2 (cis) dichlorethylene.

*Properties of a Strain-indicating Coating.*—A satisfactory strain-indicating coating should possess certain definite characteristics which are, to some extent, mutually exclusive.

(i) The lacquer must be extremely brittle and yet able to form an adherent and continuous film. The aspect of adhesion is particularly important. Certain lacquers were examined which appeared to have the requisite brittle characteristics, but could not be depended upon to give an adherent coating. As the films dried by loss of solvent, the internal stress increased, and a point was reached where they began to peel off in localized areas.

(ii) The lacquer must crack well within the elastic limit of the test material, for if it does not do so the specimen is destroyed; also the strain pattern may be materially altered as soon as plastic flow takes place in the material.

(iii) It must be capable of easy application and rapid drying. Once dry the lacquer must be permanent in its physical characteristics and should also have sufficient internal stress at this stage to prevent the cracks closing after release of the applied load.

(iv) The properties of the lacquer should, ideally, be independent of temperature and humidity change, but it is extremely doubtful whether such a material could be made which would also comply with the other requirements. Nevertheless, it should be satisfactory for the extent of variations likely to be experienced in the ambient conditions of temperature and humidity.

(v) The sensitivity should be independent of film thickness within the limits likely to be obtained by the proposed method of application.

(vi) The plasticity of the lacquer should be a minimum. If the coating is liable to plastic flow, the strain readings will vary with time of loading as the lacquer accommodates itself to the applied stress.

These were the most important characteristics of a brittle lacquer, but other features such as colour and response to dye etchants for photographic records had to be borne in mind.

*Preliminary Survey of Potential Film Formers.*—Various nitrocellulose compositions were examined in which the nitrogen content and the solvent carrier were varied. As the nitrogen content increased the films became more brittle. The films prepared from some solvents were more brittle than those from other solvents, but in no case was the promise very great, the failure being largely due to inadequate adhesion. The work was extended to nitrocellulose/gum finishes, both clear and pigmented, but here again, once the films were sufficiently plasticized to overcome spontaneous crazing and the tendency to flake, the strain sensitivity was too low. Some aspects of this work are being repeated now that controlled drying conditions are available. Cellulose acetate and allyl starch suffered from the same general type of adhesion deficiency as nitrocellulose, and were not examined further.

It was soon appreciated that a resinate type of film former offered considerable promise. This work led to the preparation of particular metallic resin complexes which, suitably plasticized with dibutyl phthalate and dissolved in carbon disulphide, form the basis of Strainlac. In many ways it was similar to Stresscoat, the American material, but differed in that it is believed that Strainlac is more sensitive to strain and less sensitive to temperature changes. The lacquers have been applied by spray either in the open air or in a conditioned air extraction booth, and dried in a double-walled Perspex cabinet which can be maintained at any temperature above the prevailing temperature. The humidity in this cabinet was controlled by various salts located in the base of the cabinet, and the method of heat transfer was by water circulated through a copper coil inside the cabinet. In view of the solvent used in these lacquers, all electrical contacts are made outside of the cabinet, the temperature and humidity being recorded on a clockwork recorder.

*Development of Resin.*—Preliminary experiments were carried out using various metallic resin complexes plasticized with dibutyl phthalate and dissolved in carbon disulphide. It was noted that different metal complexes gave different application properties; some, for example, gave continuous glossy coatings whereas others gave powdery films which were not satisfactory. Having selected the best metallic component, it was then realized that the precise metal content of the complex had a profound effect on the performance. It was therefore necessary to carry out considerable research into the effect of metal content and of the precise method

of manufacture on the sensitivity and creep characteristics of the lacquer. As a result, a rigid manufacturing schedule was laid down which specified the quality of the starting ingredients, the batch size, the heating schedule and the order and rate of addition of the ingredients to give a metal complex of the desired metal content and melting point.

The need for this close control is illustrated by the results shown in Table 1. Five samples were drawn from a proprietary batch of metallic resinate, made into lacquer, suitably plasticized and tested. The test bars were sprayed to give approximately the same depth of colour, but the film thicknesses obtained were rather outside the range which had now been adopted. Nevertheless the results in the Table show the wide variation in the constants of the samples and in the associated performance properties.

During this work an interesting observation was made by Messrs. Vickers-Armstrongs, Ltd., who had been testing some of the lacquers. A resin was prepared which was cooked for an unusually long period at high temperature with a view to increasing sensitivity. The lacquer prepared from it had given a satisfactory crack pattern when initially stressed, but had continued to crack during the next minute while under stress. This effect of "after-cracking" had not been experienced with other resins, and the reason for it was not yet fully understood; this phenomenon may be the limiting factor to sensitivity.

*Nature of Plasticizer and Solvent.*—The plasticizer should have a very low evaporation rate, and be compatible with the resin, both in the presence and absence of the solvent. Little work had been carried out to study the effect of different plasticizers, since it was felt that, provided they complied with the above requirements, the likely variation in their effect on the subsequent properties of the coating would be unimportant compared with the variation obtained from different resins. Dibutyl phthalate was found to be satisfactory, and is used in Strainlac.

The requirements for the solvent were such that the choice was very limited. It had to be highly volatile to ensure a dry film of uniform thickness; it should be pure and leave no variable "tail"; it should have no tendency to absorb moisture from the air and it should not form a complex with the resin. Small quantities of the conventional solvents, used with nitrocellulose, for example, were held very firmly by the resin for long periods after the film was dry. With the metallic

Table 1. *Metallic resinate batch No. 1*

Sample ref.	Total metal	Melting point	Mol. wt.	Film thickness	Crazing	Sensitivity	"Creep"
A	9.62	101° C	—	0.012 in	Nil	$10.0 \times 10^{-4}$	Complete
B	8.39	111° C	$1\,450 \pm 2.0\%$	0.009	Nil	8.3	Complete
C	9.44	124° C	$1\,215 \pm 1.2\%$	0.009	Considerable	—	—
D	8.34	155° C	$1\,300 \pm 1.5\%$	0.012	Nil	5.0	Nil
E	9.20	123° C	—	0.013	Moderate	—	—
Bulk	9.10	140° C	—	0.009	Slight	6.6	Nil



resinates it was found that 1% of an aliphatic hydrocarbon solvent in the mixture did not greatly affect the instantaneous sensitivity, but the cracking pattern disappeared completely on removal of the applied stress. The solvent selected for use in Strainlac was essentially carbon disulphide. This material met the essential requirements of the solvent already mentioned, and with the correct spraying techniques, gave a good glossy coating, full of very fine air bubble which appeared to help the definition of the cracking pattern. Unfortunately from other points of view, carbon disulphide was perhaps one of the least desirable solvents.

**Precautions.**—The nature of the technique of stress analysis using brittle lacquers was such that it calls for a reasonable level of intelligence by the operator, and he should be fully aware of the hazards associated with the solvent employed. Carbon disulphide was highly toxic. Table 2 gives typical figures for a few gases.

Table 2. *Toxicity of certain solvents*

Gas	Concentrations which will give rise to severe toxic effects in persons exposed to them for the stated times		Concentrations in the general atmosphere of the plant greater than those given below indicate unsatisfactory conditions	
	p.p.m.	Time of exposure	p.p.m.	
Benzene .. ..	1 500	1 h	50	
Carbon disulphide	500	1 h	10	
Chloroform ..	2 000	1 h	50	
Hydrogen cyanide	40	1 min	10	
Hydrogen sulphide	200	1 min	20	

Carbon disulphide was harmful by inhalation of the vapour, by prolonged or repeated contact of the liquid with the skin, or by oral intake. The most likely source of danger arising from the use of Strainlac was from frequent exposure to relatively low concentrations of the vapour, which, in its most serious form, results in acute mental disturbance or in severe injury to the nervous system. The other hazard associated with carbon disulphide was the fire risk. It is a highly inflammable liquid, the vapour forming highly explosive mixtures with air over a wide range of concentrations. For example, a mixture of less than 1% of the vapour in air has an ignition temperature around 81°C. As such, air/CS<sub>2</sub> mixtures can be touched off by contact with hot water or steam pipes. In view of these hazards Strainlac should either be sprayed in an efficient draught extraction booth, or applied in the open air; the operator should use the recommended type of respirator. The work on these brittle lacquers was not yet finished, and it may be possible to produce a lacquer with a less dangerous solvent.

**Method of Use of Strainlac.**—Mr. Young then described the method of use for Strainlac as follows:

- The surface of the specimen for test should be free from loose scale, oil, grease, etc.—a suitable grease solvent will be provided with the lacquers.
- A coating of a quick-drying aluminium nitrocel-

lulose lacquer is applied to the structure and the calibration bars, which are then allowed to dry for 1–2 h. This bright undercoat improves the visibility of the cracks and makes it easier to judge the thickness of the Strainlac coating by its colour.

(iii) The lacquer is then selected from the range, for the conditions of temperature and relative humidity which will obtain at the time of test.

(iv) The lacquer is applied to the calibration bars and to the test specimen from a gravity-fed spray gun using a large air volume and 5–10 lb/in<sup>2</sup> spraying pressure. The nozzle of the gun should be kept about three to four inches away from the object and an even coat built up to give a dry film thickness of 0.003–0.008 in. This may necessitate 8–10 passes of the gun over the surface. The first few passes will give a large number of discrete globules which gradually coalesce with further lacquer to give a continuous film. An operator will quickly be able to judge the coating thickness by the intensity of the yellowish green colour of the lacquer film. It will be noted that the lacquer coating contains a large number of very small air bubbles trapped beneath the top skin of resin. Table 3 indicates the effect of variation in film thickness on the sensitivity of the lacquer coatings.

Table 3. *Effect of variation in film thickness on sensitivity*

<i>Film thickness</i> <i>(in)</i>	<i>Sensitivity</i>		
0.003	$6.2 \times 10^{-4}$		
0.003	5.5		
0.004	6.2		
0.004	6.2	Lacquer ref.:	Bulk No. 3
0.004	6.0	Temperature:	62.6° F
0.005	6.0	Relative humidity:	60%
0.005	6.0	Time of drying:	16 h
0.006	5.5		
0.006	6.0		
0.007	6.0		
0.007	6.2		
0.008	6.4		
0.017	8.3		

(v) The bars and test specimen are kept together while they are dried for 12–24 h during which time the temperature and humidity should remain constant within ± 3° F and ± 5% relative humidity. Greater variations than these can be tolerated, but with the attendant risk of spontaneous crazing before test or partial “creep” after stressing.

(vi) The next step in the test is to load the test specimen. The best method depends upon the nature of the loading and the type of strain which results from it. For static loading resulting in tensile strains the method recommended is to increase the load from zero by increments with a return to zero load between each increment, until the first cracking of the Strainlac is observed. At this stage the calibration beam should be strained and the minimum strain to cause cracking noted. The position of the first cracks on the specimen should also be marked with a wax pencil before the specimen is

unloaded and the next load increment applied. Additional cracks can then be marked, until with successive increments of load an extensive strain pattern is built up. This pattern can be rendered permanently and easily visible and photogenic by using a dye etchant. This is applied by lightly rubbing the dye solution on to the surface of the lacquer film, leaving for 3–6 min, applying a second coat and then removing with a swabbing solution. The etchant colours the cracks dark red but leaves the surface otherwise substantially unaffected.

Static compressive strains could be examined by fully loading the specimen before coating and reducing the load after the drying period by successive decrements, returning to full load after each decrement.

The technique for static loading described above was used in order to reduce to a minimum the corrections required for "creep." By "creep" was meant the decrease in sensitivity which occurs if Strainlac is strained slowly. Lacquers of this type adjusted themselves to the applied load and the apparent strain indicated by cracking may be very much less than the true strain. This was because the calibration bar was in effect strained instantaneously. The return to zero load between successive increments of load on the specimen normalized the coating and so avoided the accumulation of loading times. The correction for "creep" which must be applied was that corresponding to one loading only, and was usually very small; if the loading time was constant the correction was constant. Where this technique is impracticable, full correction for "creep" could be made from a "creep" correction chart, although if long times of loading were used, sensitivity and therefore the range for strain measurement below the elastic limit of the material was reduced.

#### THE USE OF MODELS IN STRUCTURAL WORK

The morning session on Tuesday, 4 April, was devoted to three papers on this subject.

MR. E. M. LEWIS (Messrs. W. S. Atkins and Partners) described some structural engineering applications of model methods. These methods were essentially of the "indirect" type, i.e. methods depending on the measurement of deflexions at various points of a structure when a definite displacement is applied at one point. This "indirect" approach avoids the application of a definite load to the model and consequently no trouble with creep of the model material, celluloid, is encountered. By the use of the reciprocal theorem the redundant reactions of a structure can be deduced from a knowledge of the ratios of the deflexions at the points of application of the loads and reactions. Mr. Lewis described and demonstrated the type of models and apparatus he had used. The models, cut from sheets of plastic material, were geometrically similar to the structures they represented, while the widths of the members were made proportional to the cube roots of their respective second moments of cross-sectional area, so maintaining a

correct proportion between the flexural rigidities of the members. Mr. Lewis favoured methods by which the deformation applied to a structural model was large enough to be measured by simpler means than the micrometer microscopes used in the original Beggs' apparatus.

MR. J. A. H. PAFFETT (Royal Corps of Naval Constructors) discussed the use of models in the solution of shipbuilding problems. The speaker described work done by himself and his colleagues at the Naval Construction Research Establishment, Rosyth, which had led to an interesting combination of photoelastic and strain gauge methods of analysis. Complex models of parts of a ship's structure had been built up using celluloid as the model material. These, in the nature of the problem, were highly redundant three-dimensional structures.

An ingenious small portable polariscope with an outside diameter of  $3\frac{1}{4}$  in\* had been used to determine the isoclinics at a series of points on the model. From these observations a family of stress trajectories was drawn and this information was used to locate electric resistance strain gauges which could therefore be oriented in the direction of one of the principal stresses.

MR. P. J. PALMER (Boulton Paul Aircraft Ltd.) read a paper on aeronautical applications of model methods. He first discussed the suitability of xylonite as a model material. Its greatest advantage was its ease of manipulation. Owing to its thermoplastic nature it could be pressed into various shapes when hot and it could be cemented easily by a solution of 40% amyl acetate and 60% acetone. The physical properties of xylonite varied with temperature, humidity and sheet thickness and it was subject to creep, but this became very small 2 minutes after the loads had been applied. Approximate figures obtained were: limit of proportionality: 3 500 lb/in<sup>2</sup>; ultimate strength: 8 000 lb/in<sup>2</sup>; and density: 0.049 lb/in.<sup>3</sup>

Mr. Palmer also described how the "dynamic" modulus of elasticity of xylonite had been measured. If the model was to be used for both static and dynamic tests both static and dynamic model similarity must be satisfied. The non-dimensional parameters which had to be considered were

$$(1) \frac{\rho}{\gamma} \quad (2) \frac{Vl}{\nu} \quad (3) \frac{\rho V^2}{E} \quad (4) \frac{\rho gl}{E}$$

where  $E$  is modulus of elasticity,  $l$  a representative dimension,  $V$  air speed,  $\gamma$  material density,  $\nu$  kinematic viscosity of air,  $\rho$  air density and  $g$  the gravitational acceleration.

Parameter 2, Reynolds Number, could not usually be satisfied, but the effects of this were not generally serious and were largely known. Parameter 4 was the gravitational parameter which fixed the model scale, but it

\* PAFFETT, J. A. H. *Trans. N.E. Coast Inst. of Engineers and Shipbuilders.*



had been shown that, provided this factor was kept within certain limits the variation between model and full scale could be ignored. Parameter 3 determined the wind speeds; parameter 1 must also be satisfied although at first sight that seemed impossible. However, if a model was constructed true to scale with regard to external dimensions, but had the thickness of the material increased  $K$  times, the inertia and stiffness distributions of the model were identical with a geometrically perfectly similar model, but having a modulus of elasticity and density  $KE$  and  $K\gamma$  instead of  $E$  and  $\gamma$ . If  $K$  was made equal to the ratio of full-scale material density to model material density parameter 1 was satisfied. The speaker described how stiffness tests were carried out on properly proportioned models and pointed out that dial gauges could not always be used to measure deflexions as the gauge spring stiffness often affected the reading. Small mirrors afforded the best solution of this difficulty.

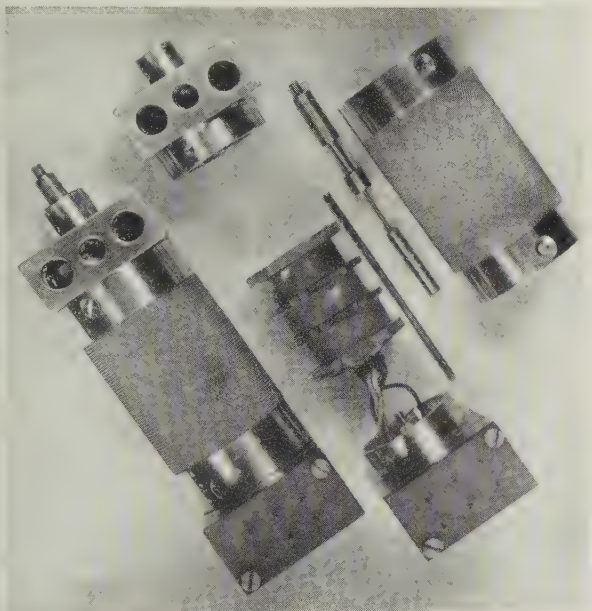


Fig. 1. Special pick up for model work

Strain gauge tests could be carried out on models in exactly the same way as on full-scale aircraft provided miniature gauges were used. The British Celanese Woven Gauge as developed by Boulton Paul Aircraft Ltd. had been used for this purpose. Model resonance tests were carried out in the same manner as full-scale tests. A mechanical oscillator was built into the model and the latter vibrated over a range of frequencies. Pick-ups were positioned on the model, so that at the resonance frequencies the amplitude, phase and frequency could be recorded.

A special pick-up for model work which is of the differential transformer type is shown in Fig. 1. The sensitivity was 20 V/in, the maximum displacement 0.1 in on either side of the balanced position. The

mass of the moving part was 5 gm and the overall length 4 in.

Several models had been built for wind tunnel measurements. Fig. 2 shows such a model which would exhibit the effects of flexibility on the aerodynamic forces.

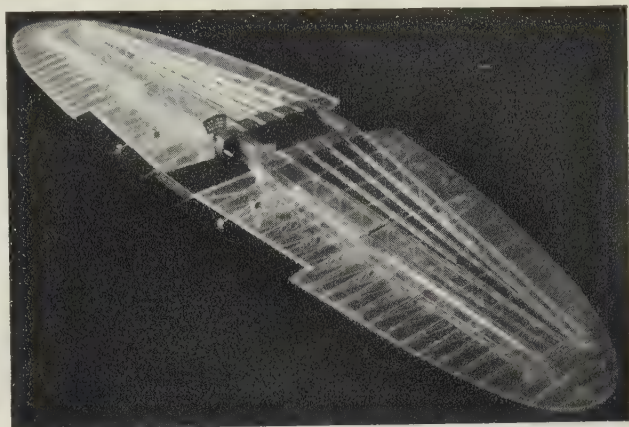


Fig. 2. Model for showing effects of flexibility on aerodynamic forces in wind tunnel measurements

#### RECENT DEVELOPMENTS IN PHOTOELASTICITY

COL. H. T. JESSOP (University College, London) described a microscopic technique for the examination of thin slices cut from photoelastic "frozen stress" models. The details of this technique have been published separately.\*

MR. H. FESSLER (Huddersfield Technical College) read a paper on "Marco Resin as a Photoelastic Model Material."

He explained that Marco Resin which is marketed by Messrs. Scott, Bader and Co. Ltd., of Kingsway, London, consists of four components which are mixed by the investigator. Tensile tests carried out on 48 different compositions had shown that the optically most sensitive composition was

Marco Resin S.B.26	..	100 parts by weight
Monomer C.	..	52 parts by weight
HCH. Catalyst	..	0.5 parts by weight
Accelerator E.	..	0.3 parts by weight

The homogeneous mixture was poured into a suitable mould and polymerized at room temperature in the absence of air and water in approximately two days. As the heat liberated in polymerization tended to accelerate the process all castings, except flat sheets, had to be water cooled.

A number of cylinders of 2 in diameter and 3-5 in long of the above composition was cast. It was found that fringes caused by the constraint of the mould disappeared rapidly after removal, but daily observations showed that the castings slowly acquired an initial stress

\* *Brit. J. Appl. Phys.*, **1**, p. 184 (1950).

of about 0.5 fr/in thickness. Heating the cylinders to 90°C for 3 h caused the stress to increase after a few days to about 1 fr/in. This was the most serious defect of Marco and attempts were being made to reduce this initial stress. It was found that all the compositions made showed rind effect, i.e. an edge stress caused by heating, of about 0.9 to 1.6 fr/in. Recovery or reduction of fringe order after "freezing" was small; weekly measurements on 21 different compositions showed a mean reduction of 2.5% over 3 months and a maximum reduction of 7%. The maximum permissible temperature which the model may reach during the slicing process was 60°C. Above that temperature the fringes disappeared rapidly. Creep tests on 12 tensile specimens of the above composition showed that at temperatures above 80°C fringe order and elongation reached constant values in less than 1 h after application of the load. Five specimens reached their full elongation in less than 4 min. For freezing temperature of 80°C to 110°C, these specimens showed the following mean residual values:

Material fringe value ( $f$ )	..	4.7 lb/in/fr
Young's modulus $E$	..	2 800 lb/sq in
Optical modulus ( $O$ )	..	600 fr/in

where  $O$  is the number of fringes which an ideal specimen 1 in thick would show if elongated by its original length.

MR. P. GRODZINSKI (Diamond Research Department, Industrial Distributors, Ltd.), read a paper on "Photoelastic Investigations on Diamond Crystals." He pointed out that interest in the stress-optical properties of diamond was not new. G. Wertheim in 1854 had measured the stress-optical constants of various substances, amongst them diamond, for which he advanced a stress optical coefficient  $C = 2.318$  in metric units and also gave a modulus of elasticity of 10.865 kg wt/mm<sup>2</sup>. Later F. Pockels had shown that even in cubic crystals like the diamond which is singly refractive, the apparent stress-optical coefficient is a function of orientation. Wertheim had not stated how his crystals were cut with reference to their axes of symmetry (mentioned by Coker and Filon).

Little or no use had been made in practical stress-optical investigations of the highly developed general theory of photoelasticity in crystals. This general theory of photoelasticity was developed by F. Pockels and in 1935 a new theory was advanced by H. Mueller. Pockels had determined the photoelastic constants of various cubic crystals, but not that of diamond. Recently G. N. Ramachandran had recapitulated the general theory of photoelasticity for cubic crystals and had determined for the first time the photoelastic constants of diamond on three diamond specimens, stressed in compression. The values had been checked by E. Burstein and P. L. Smith in 1948, and some changes had been made in agreement with Ramachandran. Such crystals besides diamond for which the photoelastic constants had been determined and which might be of

interest for stress-optical investigations are: Magnesium oxide, lithium fluoride, potassium bromide, silver chloride, thallium chloride bromide, thallium bromide iodide and zinc sulphide. These materials he gathered had been used for photoelastic purposes by G. Rosenberg and A. V. Stepanov. Sir William Crookes had advanced the opinion that the majority of diamonds showed strain, usually centred about an inclusion or flaw.

Rough diamonds used for dies have been inspected under the microscope in polarized light in order to find out whether they were under stress. Diamonds which showed a stress pattern were subsequently rejected. No attempt was made to determine the magnitude of the stress. At the time (1934) the method seemed to be a quite suitable one, as an existing stress pattern may have made the diamond unsuitable for the use as a die. Later on it was realized, however, that the diamond used as a tool must be able to sustain quite appreciable stresses starting at the points where it comes into contact with the work, and at those points where it is clamped to the holder. In the literature quite a number of instances had been reported of rough diamonds exploding without any explanation being advanced. This was probably due to the release of internal strain. Again, in one instance a diamond was bruted, i.e. worked with another diamond, and had thus undergone severe treatment. Soon afterwards it was reported that the diamond had been shattered without being stressed.

Whilst the investigation of strain and stress in natural stones seemed of somewhat limited scope, as the extent of the stress could not be determined with any degree of accuracy, the photoelastic investigation of actual tools seemed to open an interesting aspect of the subject. For instance, in diamond dies and generally in dies, it was important that the casing exert a certain pressure which was released during the actual drawing process. Jearum and Grodzinski in British Patent 549407 (1941), suggested observation through the windows usual with diamond dies of the stress in the die before, and probably during the process. Coker and Filon had originally suggested a model mechanism for investigating the stresses in both the cutting tool edges and the chip.

In a recent investigation of the cutting conditions of diamond tools by polarized light, a simple apparatus (designed by P. H. Rinkel) which was mounted on the table of an inverted microscope. The edge of a plane parallel diamond (similar to a grooving tool) could be stressed by a plate and the pressure measured by a spring dynamometer. When the diamond was viewed stress patterns were observed near the point, as well as a more intense stress pattern where the tool was clamped. It had not yet been possible to determine the actual stresses owing to the complicated nature of the diamond crystal. The basic idea of this set-up was to use the diamond as a kind of dynamometer by observing its stress pattern. Any other kind of dynamometer would only measure the average cutting pressure, whereas in this instance the



pressure distribution over the tool tip could be followed up in detail. Such a study might give information on the suitability of certain tool edges and indicate how they could be improved. As no suitable data were available in the literature for the photoelastic constants of diamond, new investigations were made and W. A. P. Fisher designed an apparatus for determining the stress-optical coefficient of small diamond needles of about 1 mm<sup>2</sup> cross-section and 5 mm length subjected to pure bending. One long face of this needle was a natural octahedron face and (111) the other face was perpendicular to it; (112), the principal stress was directed along the zone axis.

The following values had been obtained for the stress optical coefficient in Brewsters:

Needle				(111)	(211)
No. 0	..	..	..	7.15	8.2
No. 1	..	..	..	9.45 (12.2)	7.4
No. 2	..	..	..	8.75 (6.9)	9.35
Averages	..	..	..	8.45 $\pm 1.0$ - 1.35	8.32 $\pm 1.0$ - 0.9

Calculated from experimental data (Ramachandran, 1947)*	5.48	1.83
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\* Calculated by Dr. W. A. Wooster.

Judging from these experimental data the two different planes give practically the same photoelastic constants. There was, however, the great discrepancy between the values found by Ramachandran and those obtained in this investigation which had not yet been explained. On the assumption that the stress optical coefficients in both directions were the same, the method already developed could be usefully employed for the actual determination of the stresses in diamond tools.

Enlarging further on the use of diamonds the speaker suggested the use of such crystals as optical dynamometers, replacing the use of piezo-electric apparatus: thus it would be possible to measure the actual magnitude and distribution of the stresses instead of average pressures as given by piezo-electric crystals.

#### APPLICATION OF EXPERIMENTAL STRESS ANALYSIS METHODS TO SOIL MECHANICS PROBLEMS

MR. A. A. WELLS (Cambridge University Engineering Department) read a paper on "Strain Gauge Techniques in Soil Dynamics." He described the use of strain gauges for the measurement of the forces acting on the component parts of certain type of ploughs. For the simultaneous recording of dynamic strains at moderate frequency response, without amplification, the speaker had used a multi-channel string galvanometer because of its simplicity, robustness and portability. A fourteen-string galvanometer, with permanent magnet, a natural frequency of 70 c/s, full-scale deflexion  $\pm 40$  A, a string resistance of 12  $\Omega$  and critical damping with an external

circuit resistance of 200  $\Omega$  was used under severe field conditions.\*

MR. J. C. HAWKINS (National Institute of Agricultural Engineering) spoke next on "A Strain Gauge Dynamometer Rig for the Determination of Soil Force on Plough Bodies." He explained that previous investigators had used hydraulic dynamometers but that at N.I.A.E. they had decided to use electric resistance strain gauge dynamometers for a variety of reasons. The main advantages of the electrical dynamometer were considered to be their great sensitivity to transient forces, their much smaller bulk and the fact that signals could be more easily and conveniently conducted along a multi-core cable than along a multiple hydraulic line.

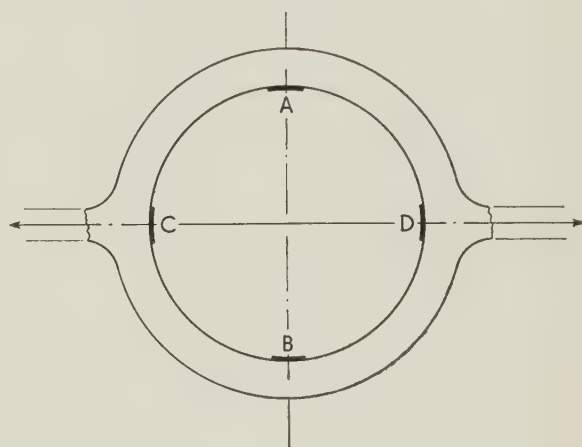


Fig. 3. Showing position of strain gauges in ring dynamometer

Fig. 3 shows the dynamometer diagrammatically. This type of dynamometer was suggested to the N.I.A.E. by the late Mr. Eric Jones of the Royal Aircraft Establishment. In its simplest form four gauges are used and are bonded to the inside of the ring at four points on mutually perpendicular diameters. Gauges at *A* and *B* are in tension for a tensile load on the ring and those at *C* and *D* are in compression. The distribution of stress round the ring is as shown in Fig. 4. The intensity of compressive stress at *C* and *D* is higher than at *A* and *B* but extends over a smaller circumferential length. It was therefore important to use gauges of as short a gauge length as possible.

In the rings which were manufactured for N.I.A.E. a total of eight gauges was specified, two gauges being stuck in each position side by side so as to behave as one wide gauge. The main reason for bonding two gauges in each position was so that any bending of the ring inadvertently transmitted could be cancelled out by suitable cross-connexion of the gauges. By this means every secondary effect which occurred in any one arm of

\* *J. Sci. Instrum.*, 27, p. 59 (1950).

the Wheatstone bridge was wholly balanced by an identical one in an adjacent arm. The method is shown in Fig. 5. For clarity the gauges are represented as one on top of the other although they are actually side by side. The rings were made of copper-beryllium. This

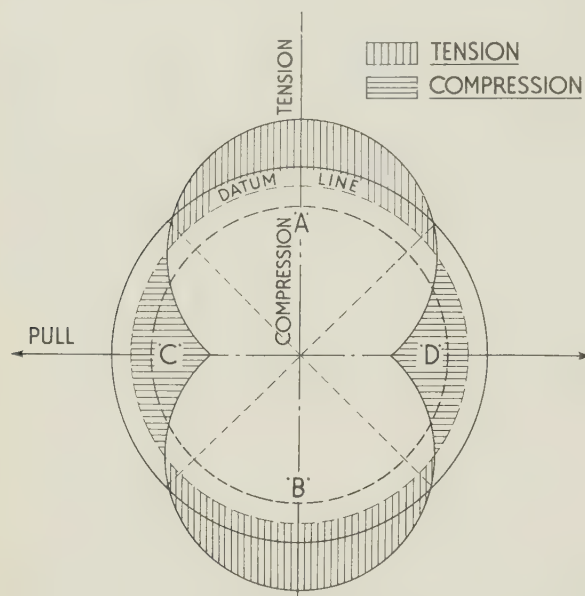


Fig. 4. Stress distribution on inside face of a ring under diametral tension

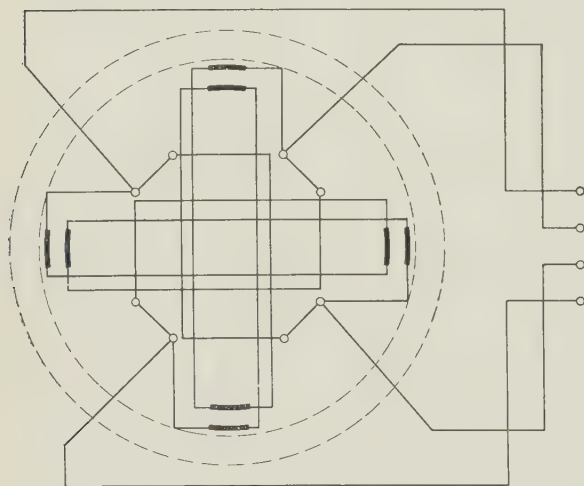


Fig. 5. Connexion of eight strain gauges in ring dynamometer

material was chosen for its high proof stress and low modulus of elasticity, which made the dynamometer nearly twice as sensitive as a similar high-grade steel ring having the same factor of safety. 120  $\Omega$  gauges by Baldwin Southwark were bonded to the metal by means of a thermoplastic cement and the leads were taken

from the gauges separately to a terminal board to which outside leads could be connected.

It was necessary to make sure that the rings were entirely weatherproof and for this reason the rings were first of all baked out, then Di-jel 171 was poured in the rings, which were finally sealed off with rubber bonded on to the metal with bostic. The terminals for the leads were bonded into perspex sheet bonded in turn to the metal with bostic. The whole of the ring was encased in bakelized fabric machined to shape and varnished. The sensitivity of the ring was very much greater than that which could be obtained by using a flat bar or strip for the same rigidity. Because all four arms of the bridge were within an inch or two of each other they were subjected to identical temperature changes and the problem of temperature sensitivity was very much eased. The gauges were, moreover, heavily heat insulated by the mass of Di-jel in the rings and therefore there was no question of stray draughts affecting one or more of the gauges. They had found that a short warming-up period was necessary while a small amount of initial drift occurred, presumably due to differences in thickness of the adhesive between each gauge and the metal, but equilibrium was attained after a few minutes and there was no further drift for long periods. The maximum loads for the dynamometers used up to the present ranged from 500 to 2000 lb. The bridge current used was 50 mA, giving a current per gauge of 25 mA. Using galvanometers of about 520  $\Omega$  resistance the sensitivities ranged from 0.3 to 0.6 mA/10 lb. It was proposed to employ two four-channel dynamic strain-recording units by Henry Hughes & Son, Ltd., incorporating dry ink recorders to record the readings from the dynamometer rings. Mr. Hawkins also described the recording system in temporary use at the present time until delivery of the recorders was obtained.

MR. A. W. BISHOP (Imperial College of Science of Technology, London) described a new method for the laboratory measurement of the coefficient of earth pressure at rest ( $K_0$ ). The property was of importance in the design of rigid underground structures and in the study of the shear strength of naturally deposited soils. It is defined as the ratio of the horizontal to the vertical effective stresses in a natural or artificial deposit in which lateral deformation is prevented. Several laboratory methods had been tried in the past, the two main types being those used by Kjellman in 1936 and Tschebotarioff in 1949, in which the sample was contained in a rigid box or cylinder (and was therefore subject to vertical shear forces on its boundaries during loading); and that used by Gersevanoff in 1936, in which the sample was placed in a rubber membrane and lateral yield largely prevented by enclosing it in a sealed vessel full of water. For sands the results obtained varied from  $K_0 = 0.5$  (Kjellman and Tschebotarioff) to  $K_0 = 0.41 - 0.36$  (Gersevanoff).

The apparatus used by Mr. Bishop for saturated samples of sand is illustrated in Fig. 6. A cylindrical



ample is enclosed in a rubber membrane and connected through a porous disk at its base to a burette. An axial strain is applied to the sample by means of the ram, and the cell pressure is controlled by a screw-operated piston in such a way as to maintain a constant cross-section in the sample. This condition is satisfied when the volume of water expelled from the sample corresponds to the axial deformation multiplied by the initial cross-sectional area. The slow rate of loading permits this to be maintained to a close approximation throughout the

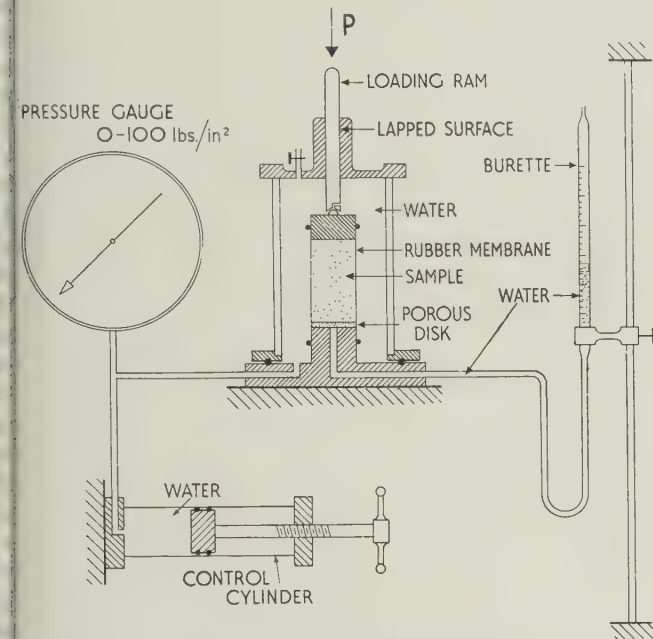


Fig. 6. Showing layout of apparatus for measuring  $K_0$  with saturated samples

test by manual control. The use of a thin rubber membrane ensures absence of vertical shear on the side of the sample. For a clean medium sand the results indicate values of  $K_0$  ranging between about 0.45 for loose packing to 0.35 for dense packing. These are average results for a pressure range of 0 to 200 lb/in<sup>2</sup>, the value of  $K_0$  tending to rise slightly with increasing pressure. Tests have been carried out on dry sand by a similar but more elaborate method. It is hoped to publish fuller details of the technique and results at a later date.

MR. R. G. BOITEN (Delft, Holland) described a number of instruments incorporating electric resistance strain gauges which had been used in soil mechanics investigations in Holland.

#### STRESS ANALYSIS IN THE MOTOR INDUSTRY

At the afternoon session on 5 April two papers were presented which dealt with the use of experimental stress analysis methods in the motor industry.

VOL. 1, OCTOBER 1950.

MR. R. S. BENNETT (B.S.A. Group Research Centre) read a paper on "A Portable Strain Gauge Equipment for use on Motor Cycles." The author pointed out that the problem of measuring and recording dynamic strains on a motor-cycle was far more difficult than in the case of a motor-car. The considerable vibration and lack of space tended to cramp the style of the designer so that any controls or refinements which were not absolutely essential had to be dispensed with for the sake of simplicity and portability.

Resistance wire strain gauges had been chosen for reasons of adaptability, cheapness and good accuracy of measurement, and of the two methods of using strain gauges for dynamic measurements, i.e. in either a d.c. or a.c. bridge circuit, the latter was chosen to ensure a good low frequency response and to provide a simple means of measuring the vibration frequencies. The a.c. carrier frequency method also overcame the difficulty of constructing a good d.c. amplifier. They had purposely limited themselves to a single channel equipment and the recording was obtained on a 16 mm film, being a photograph of the light spot on a miniature cathode-ray tube. The schematic diagram of the whole equipment is shown in Fig. 7.

Fig. 8 shows the circuit diagram of the oscillator unit, which consists of four miniature pentodes type 8D3. The first two valves  $V_1$ ,  $V_2$  form a phase-shift oscillator with a three section CR feed-back network operating at

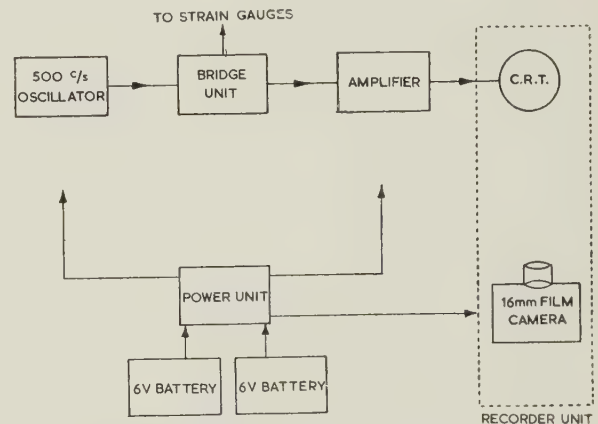


Fig. 7. Schematic diagram of layout of portable strain gauge equipment for use on motor-cycles

a frequency of 500 c/s. This frequency was chosen as being the minimum to enable vibration frequencies up to about 50 c/s to be recorded satisfactorily. The second valve  $V_2$  is also a phase splitter and the outputs from its anode and cathode are used to drive the push-pull output stage formed by valves  $V_3$ ,  $V_4$ . A maximum undistorted output of just over 2 W is obtainable when supplying a 1 000  $\Omega$  gauge bridge circuit. This is equivalent to a gauge current of approximately 22 mA. The bridge circuit consisted essentially of a simple Wheatstone bridge with equal resistance arms, but since it was

operating at 500 c/s, it was necessary to provide a capacity phase balance as well as the ordinary resistance balance. Coarse and fine resistance balance controls were provided, the latter being a helical type potentiometer of total resistance  $20\ \Omega$  limited by a smaller resistance of  $5\ \Omega$ . A ten-position switch formed the coarse capacity balance and there was also a small variable air condenser in parallel as a fine control. A two-way toggle switch enabled the capacity to be applied to either side of the bridge. The amplifier (see Fig. 9) consisted of three stages of pentodes, types 8D3, and

should not prove a serious drawback. Four miniature 90 V batteries were used to provide the h.t. supplied to the amplifier and oscillator, the actual applied h.t. being adjustable by means of two preset resistance controls. The h.t. for the cathode-ray tube was obtained from a special miniature 300 V battery, and two 6 V motor-

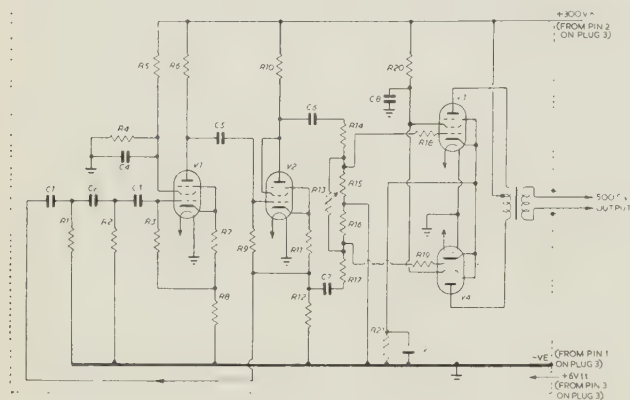


Fig. 8. Oscillator unit

## Component Values

$R_1$ 166 k $\Omega$ (HS)	$R_{11}$ 2.2 k $\Omega$	$C_1$ 500 $\mu$ F (SM)
$R_2$ 250 k $\Omega$ (HS)	$R_{12}$ 47 k $\Omega$	$C_2$ 500 $\mu$ F (SM)
$R_3$ 250 k $\Omega$ (HS)	$R_{13}$ variable	$C_3$ 500 $\mu$ F (SM)
$R_4$ 33 k $\Omega$	$R_{14}$ 470 k $\Omega$	$C_4$ 0.01 $\mu$ F
$R_5$ 150 k $\Omega$	$R_{15}$ 68 k $\Omega$	$C_5$ 0.001 $\mu$ F
$R_6$ 68 k $\Omega$	$R_{16}$ 68 k $\Omega$	$C_6$ 0.001 $\mu$ F
$R_7$ 750 $\Omega$	$R_{17}$ 470 k $\Omega$	$C_7$ 0.001 $\mu$ F
$R_8$ 680 $\Omega$	$R_{18}$ 10 k $\Omega$	$C_8$ 0.01 $\mu$ F
$R_9$ 1 M $\Omega$	$R_{19}$ 10 k $\Omega$	$C_9$ 100 $\mu$ F
$R_{10}$ 47 k $\Omega$	$R_{20}$ 3.3 k $\Omega$	
	$R_{21}$ 150 $\Omega$	

included frequency selective negative feedback to provide a reasonably flat response between 450 and 550 c/s, at the same time attenuating all other frequencies. In this way the noise level due to microphony, etc., was reduced to a minimum, and a maximum overall gain of about 100 000 at 500 c/s was obtainable. A double gang potentiometer in the input circuit provides a variable gain control. The recorder unit included a  $1\frac{1}{2}$  in diameter cathode-ray tube and a modified ex-R.A.F. camera type G45B for photographic recording. A voltage of only 300 V applied to the cathode-ray tube had been found to provide satisfactory brilliance and also increased the deflexion sensitivity. The film-drive mechanism had been modified to give a continuous film speed of 5 in/sec in order that a record of the full 500 c/s waveform will be obtained, thus providing a simple means of checking the modulation frequencies. The main disadvantage of this system was the large quantity of film required, but it obviated the need for a timing oscillator to give calibration pips on the film, and if used with care

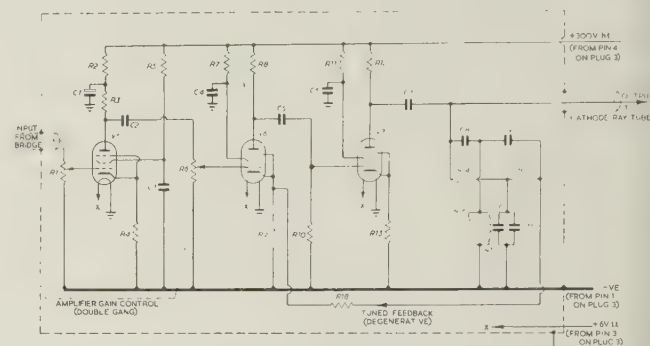


Fig. 9. Amplifier unit

## Component Values

$R_1$ 1 M $\Omega$	$R_{10}$ 470 k $\Omega$	$C_1$ 2 $\mu$ F
$R_2$ 10 k $\Omega$	$R_{11}$ 270 k $\Omega$	$C_2$ 0.01 $\mu$ F
$R_3$ 100 k $\Omega$	$R_{12}$ 100 k $\Omega$	$C_3$ 0.01 $\mu$ F
$R_4$ 470 $\Omega$	$R_{13}$ 2.2 k $\Omega$	$C_4$ 0.01 $\mu$ F
$R_5$ 680 k $\Omega$	$R_{14}$ 470 k $\Omega$	$C_5$ 0.01 $\mu$ F
$R_6$ 1 M $\Omega$	$R_{15}$ 470 k $\Omega$	$C_6$ 0.01 $\mu$ F
$R_7$ 220 k $\Omega$	$R_{16}$ 470 k $\Omega$	$C_7$ 0.01 $\mu$ F
$R_8$ 68 k $\Omega$	$R_{17}$ 470 k $\Omega$	$C_8$ 0.001 $\mu$ F
$R_9$ 680 $\Omega$	$R_{18}$ 47 k $\Omega$	$C_9$ 0.001 $\mu$ F
		$C_{10}$ 0.001 $\mu$ F
		$C_{11}$ 0.001 $\mu$ F

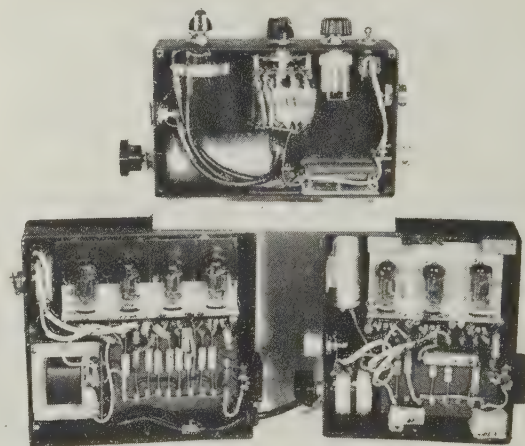


Fig. 10. Oscillator, bridge unit and amplifier

cycle accumulators provided the necessary power for the valve heaters and camera motor. All the various voltages could be monitored on a voltmeter by operation of a selector switch.

The oscillator, bridge unit and amplifier had been constructed in the form of three separate aluminium containers to facilitate screening and portability. They are shown with their covers removed in Fig. 10. The



total weight of these three units is less than 7 lb. The oscillator valves and components were mounted on a thin paxolin panel which was supported by means of four extension springs to the inside of the container. The amplifier was similarly constructed, but it was found better to mount this panel of components on small pads of sorbo rubber to reduce the effects of external vibration. It was hoped to carry these three units in a haversack container attached to the rider's back. The recording unit consisted of the cathode-ray tube and the camera, both attached to a paxolin board. It was felt that this unit could be made more compact than at present by use of a prism or mirror system, but even so it was hoped to be able to carry out some tests with this unit strapped along the top of the petrol tank. The weight of this unit is about 12 lb. This equipment had not yet been used on road tests, but very satisfactory results had been obtained on bumper tests in a factory.

The noise level for maximum gain was equivalent to an input voltage of  $4\text{ }\mu\text{V}$  and thus formed no limiting factor to the minimum strain which could be measured. In spite of the balancing controls of the bridge circuit, however, there was always a small out-of-balance voltage which was equivalent to a strain of about  $3 \times 10^{-6}$ . It is thus quite possible to detect strains as low as this, although measurements would not be reliable for strains less than about  $10 \times 10^{-6}$ . As far as microphony of the valves was concerned they did not anticipate much trouble from this source provided the amplifier and oscillator units were packed with sorbo rubber in a suitable carrier or carried on the rider's back. Movements of the amplifier unit did not increase the general noise level, unless the casing was tapped by direct contact.

The conference closed with a lecture by Mr. L. F. ATKINSON (Research Dept., Vauxhall Motors Ltd.) on "Experimental Stress Analysis Methods in the Motor Industry." Mr. Atkinson outlined the history of the use of such methods in the industry from carbon resistance strain gauges onwards. He had used brittle lacquer techniques for a considerable time, beginning with the use of ordinary "plumber's resin." This had been found too difficult to apply evenly to

complex parts and after trying a number of different lacquers "Stresscoat" had been adopted for his work. Vauxhall Motors Ltd. had installed a satisfactory spray booth and the technique of application was the standard procedure recommended by the makers. The interpretation of results was based on the "first-crack" method only if the test was carried out under laboratory conditions. When tests were carried out on the road the author has obtained satisfactory, though admittedly less accurate results by comparing the spacing of the crack lines on the part under investigation with the spacing obtained on the standard cantilever calibration bar.

After illustrating the use of Stresscoat by a number of practical examples Mr. Atkinson turned to the use of strain gauges. He used cellulose bonded gauges fairly extensively for static tests and tests under laboratory conditions, but resin bonded gauges more extensively on road tests. For all road work and wherever gauges were in use near oil or water bakelite cement type NPA had been found a satisfactory adhesive. Mr. Atkinson explained how strain gauges had been used to solve a number of difficult stress problems in his work. He outlined the construction of a towbar unit which had been found useful to measure the pull exerted by a motor vehicle. The towbar was a hollow tube with an eyebolt at each end. Strain gauges were stuck face down on to a transfer paper, two arms axially and two circumferentially; the circumferential gauges took advantage of the lateral compression and provided temperature compensation. The transfer was wrapped round the tube which had been treated with Bakelite cement and then bound with thread in a lathe (like a cricket bat handle). The whole assembly was then baked and a final layer of porous bandage was put on with more bakelite cement and baked to give a hard, oil and waterproof surface. All plug connexions were waterproofed with Di-jel 171.

The lecturer also explained the use of an axle torque-meter employing electric resistance strain gauges, which, mounted on a car, was demonstrated after the lecture.

E. K. FRANKL

## ORIGINAL CONTRIBUTIONS

# Propagation of Low Frequency Ultrasonic Waves in Rubbers and Rubber-like Polymers

By P. HATFIELD, M.Sc., A.Inst.P., Dunlop Rubber Co., Ltd., Birmingham

[Paper received 30 June, 1950]

The measurement of the velocity and absorption coefficient of low amplitude ultrasonic waves in natural and synthetic rubbers, and thermo-plastics is described. The experiments cover the frequency range of 50 to 350 kc/s and a temperature range 0 to 60° C. It is found that the velocity varies from  $1.1 \times 10^5$  to  $1.8 \times 10^5$  cm/sec at room temperatures and the absorption varies from less than 0.1 to 2 db/cm at 50 kc/s frequency and from 0.7 to 11 db/cm at 350 kc/s in different rubbers. The applications of these materials as lenses for focusing and media suitable for the transmission of ultrasonic waves are discussed, along with calorimetry.

The practical application of ultrasonic techniques for finding internal air flaws in rubber has been investigated, and to help this work it was essential to measure the acoustic resistivity (velocity of wave in rubber multiplied by specific gravity of rubber) and absorption of ultrasonic waves in a few of the many available rubbers. Experiments have been made to measure the velocity and absorption of ultrasonic waves in rubbers in the lower ultrasonic frequencies, 50 to 350 kc/s, as these frequencies are the most suitable for internal air fault detection. The aim of these experiments has been to compare the velocity and absorption of ultrasonic waves in different rubbers with reasonable accuracy. Most of the work has concentrated on natural rubber, and natural rubber compounded with different percentages of carbon and iron oxide fillers. Also included are results for common synthetic rubbers, such as neoprene, hycar, butyl, and perbunan, and also the thermo-plastics, polystyrene, polyvinylchloride (p.v.c.), polymethylmethacrylate (Perspex), and polythene. The velocity of the ultrasonic waves in rubber samples is very similar to the value in water of  $1.5 \times 10^5$  cm/sec; the specific gravity of the samples is about unity. It is therefore possible to select materials which can be used as transmission plates, or as acoustic lenses for different liquids. Some of these materials dissolve in certain liquids, but it should be possible to select from this wide class of rubbers and plastics, a suitable material for ultrasonic work.

## VELOCITY MEASUREMENTS

The velocity of small amplitude longitudinal bulk ultrasonic waves in rubber was measured by a method similar to Klein and Herschberger's.<sup>(1)</sup> It consists of finding the phase difference between an ultrasonic wave passing through water and one through rubber. The phase differences are recorded on a cathode-ray tube. The method is only applicable to solids which have an acoustic resistivity similar to that of a liquid. Fortunately most rubbers are in this category. If the acoustic resistivity of the solid differs appreciably from the liquid, then waves incident on a liquid-solid boundary are reflected, and the accuracy of the method is reduced. The block diagram of the method is shown in Fig. 1. An electrical oscillation is fed by co-axial cable to a

transmitting X-cut quartz crystal immersed in a water-filled interferometer, and a variable voltage is fed to the X-plates of a cathode-ray oscilloscope. The frequency of the electrical oscillator is the same as the resonant frequency of the quartz crystal. Ultrasonic waves are transmitted from the quartz crystal, and are picked up by a similar quartz receiving crystal parallel and near to the transmitting crystal. The receiver crystal converts the ultrasonic waves into a small electrical oscillation, which is amplified and then fed to the Y plates of the cathode-ray oscilloscope. The two alternating voltages on the X and Y plates combine to form a simple Lissajous figure. The receiving crystal is connected to a long 2.5 cm diameter lead screw, and can be moved to and from the transmitting crystal. The voltages on the X and Y plates are adjusted to be approximately equal.

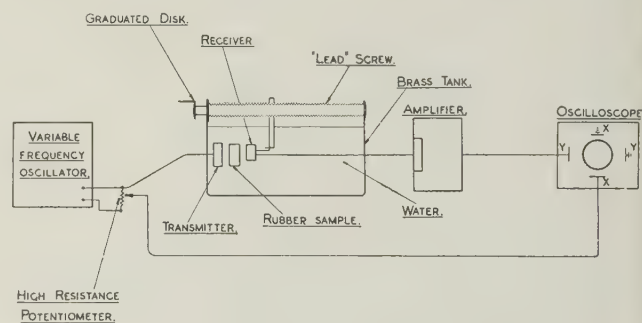


Fig. 1. Block diagram of apparatus

Moving the receiver away from the transmitter gives an ellipse on the oscilloscope which changes to a straight line inclined at approximately 45° to the X axis, grows to an ellipse again, and then to another straight line at approximately 135° to the X axis. The distance moved by the receiving crystal to produce two successive straight lines is equal to half the wavelength of the ultrasonic waves in the water. To measure the velocity of ultrasonic waves in rubber, the transmitter and receiver are set close together and adjusted to give a straight line position on the oscilloscope. Inserting a known thickness of rubber between the receiver and transmitter causes the straight line to change to a narrow ellipse; moving the receiver causes this ellipse to return



to the straight line position. The velocity of sound in the rubber can be calculated from the following formula:—

$$c_r = c_w t / (t \pm x)$$

Where

$c_r$  = velocity of ultrasonic wave in rubber.

$c_w$  = velocity of ultrasonic wave in water.

$t$  = thickness of rubber.

$x$  = distance receiver is moved after inserting the solid. The sign of  $x$  depends on whether the receiver moves towards or away from the transmitter.

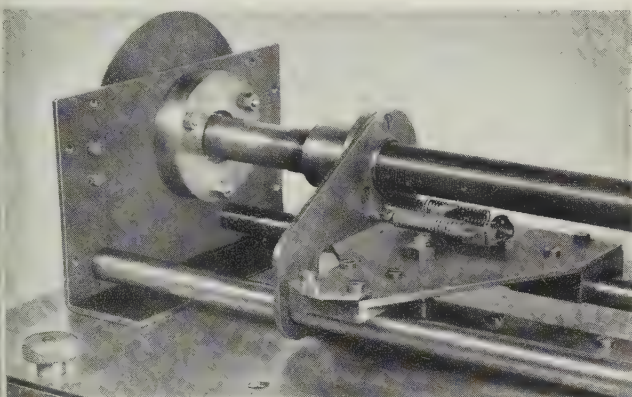


Fig. 2. Top section of acoustic interferometer

Fig. 2 shows details of the brass interferometer  $80 \times 80 \times 40$  cm. A long 2.5 cm diameter mild steel lead screw carries a collar and platform, to which is attached the receiver. The pitch of the lead screw is 0.1 cm, and a large dial divided into 100 divisions is fastened to one end of the lead screw. The collar and platform are separate but are joined by a small spring loaded rod. This allows the receiver and platform which slides on two 1.2 cm parallel rods, to move in a straight line, whereas the collar may move with a corkscrew motion. Identical X-cut quartz crystals of 2.5 cm diameter were mounted in brass crystal holders, one pair resonated at 50 kc/s frequency, and the other pair at 300 kc/s. The rubber samples used were normally  $5 \times 5 \times 2.5$  cm. If the absorption was very high thinner samples had to be used. If the samples were thicker than 2.5 cm spurious side reflexions were picked up by the receiver.

The velocity of the ultrasonic wave was measured in a large number of typical rubber compounds at 50 kc/s frequency and at room temperature. These results are given in Table 1. From this table, it will be seen that rubber can be compounded to have, at room temperature, an acoustic resistivity similar to water, and by suitable addition of compounding ingredients, the acoustic resistivity can be made to vary. Also, the ultrasonic velocity in different rubbers may be greater or less than the velocity in water. Fig. 3 shows the variation of velocity of the ultrasonic waves in rubbers by the addition of iron oxide. The effect on the velocity of waves in the rubber is very marked. The addition of

Table 1. Ultrasonic properties of rubber  
Frequency 50 kc/s. Temperature 17°C.

	Ratio by wt. of rubber to filler	Velocity cm/sec.	Specific gravity	Acoustic resistivity $\rho$ C cm/sec.
<i>Natural rubber base.</i>				
(1) Uncured pure rubber	—	$1.60 \times 10^5$	0.986	$1.58 \times 10^5$
(2) Rubber plus only 10% sulphur	—	1.60	0.995	1.59
(3) Rubber/carbon	100/47	1.61	1.11	1.79
(3a) Rubber/carbon	100/140	1.68	1.31	2.20
(4) Rubber/ironoxide	100/54	1.38	1.31	1.81
(5) Rubber/ironoxide	100/108	1.29	1.60	2.06
(6) Rubber/ironoxide	100/216	1.24	2.03	2.52
(7) Rubber/ironoxide	100/270	1.21	2.25	2.72
(8) Rubber/china clay	100/27	1.52	1.13	1.72
(9) Rubber/china clay	100/136	1.44	1.45	2.09
(10) Rubber/magnesium oxide	100/119	1.50	1.34	2.01
(11) Rubber/silene	100/23	1.52	1.05	1.60
(12) Rubber/silene	100/66	1.53	1.21	1.85
(13) Rubber/silene	100/114	1.56	1.37	2.14
(14) Rubber/litharge	100/360	1.00	3.34	3.34
(15) Ebonite	—	2.50	1.26	3.15
Water	—	1.50	1.0	1.50
<i>Synthetic rubber base.</i>				
(16) Neoprene/carbon	100/14	1.58	1.30	2.05
(17) Neoprene/carbon	100/60	1.69	1.42	2.40
(18) Hycar O.R./carbon	100/6	1.76	1.06	1.87
(19) Hycar O.R./carbon	100/58	1.74	1.16	2.02
(20) Perbunan/carbon	100/7	1.63	1.07	1.74
(21) Perbunan/carbon	100/58	1.68	1.18	1.98
(22) Butyl/carbon	100/40	1.60	1.10	1.76
<i>Plastics.</i>				
(23) Polythene	—	1.7	0.99	1.68
(24) Polyvinylchloride	—	2.3	1.50	3.45
(25) Perspex	—	2.0	1.2	2.40
(26) Polystyrene	—	2.5	1.02	2.55

Note samples 3–14, 16–22 had in addition small percentages of compounding ingredients—sulphur 3 parts, stearic acid 1, and M.B.T. 0.85. Sample 24 contained 30% by weight of plasticizer dioctyl phthalate.

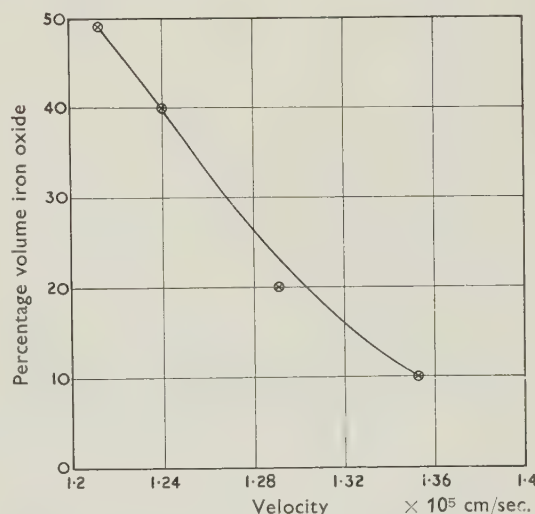


Fig. 3. Variation of ultrasonic velocity in natural rubber with addition of iron oxide. Frequency 50 kc/s

litharge and zinc oxide in large quantities similarly reduced the velocity of the wave in rubber. Only one value for ebonite is quoted but this material depending on the compounding ingredients gives a wide velocity

variation. The velocity of sound in the synthetic rubbers is usually slightly higher than the velocity in natural rubber, but the velocity in neoprene and butyl rubbers is similar to that of natural rubber. The addition of carbon black tended to increase slightly the velocity of ultrasonic waves in rubber. The number of different carbon blacks is extremely large but samples of different blacks gave similar results. Fig. 4 shows typical curves for the variation of the velocity of the ultrasonic wave in the rubber with temperature at a frequency of 50 kc/s. A very rapid rise in ultrasonic velocity occurs as the

$\times 10^5$  cm/sec.

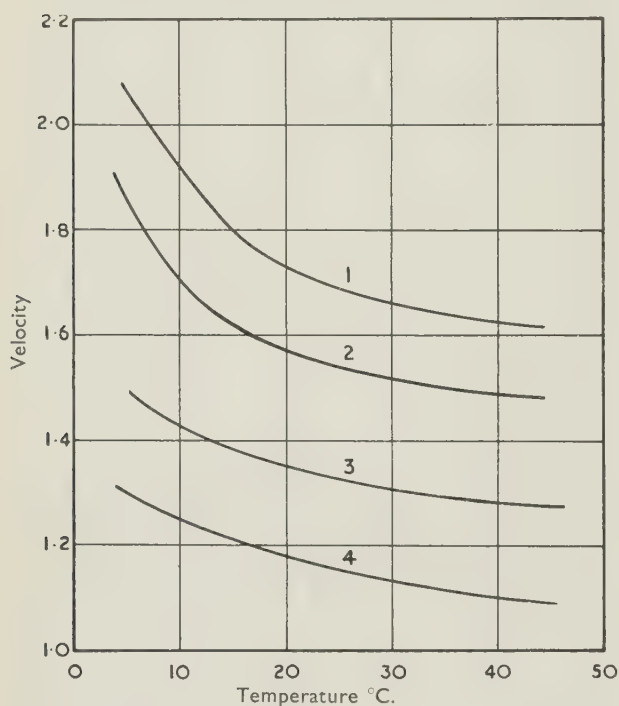


Fig. 4. Velocity  $v$ . temperature curves for different rubbers at 50 kc/s

- (1) Hycar 6% by wt. carbon.
- (2) Natural rubber 47% by wt. carbon.
- (3) Natural rubber 54% by wt. iron oxide.
- (4) Natural rubber 270% by wt. iron oxide.

temperature of the rubber approaches  $0^\circ\text{C}$ . The experiments have been repeated at 75, 150, and 300 kc/s and the velocities at the highest frequency tended to be 3% higher than the lowest frequency.

A number of errors are associated with the experiment, particularly at the lowest ultrasonic frequency. The transmitted waves are not plane and reflexions from the sides of the tank can be picked up by the receiving crystal. Working with the transmitter and receiver as close together as possible reduces this effect. Another error is caused by minute air bubbles on the surface of the rubber and it is advisable to soak all rubber samples for 24 h in water before making measurements. It is also essential that air is not trapped in the moulded rubber samples, and it is advisable to mould several samples of the same rubber.

## ABSORPTION MEASUREMENTS

Ultrasonic absorption measurements were made by sending an ultrasonic wave through the rubber sample immersed in water and measuring the voltage drop on a voltmeter coupled to the receiving crystal and comparing this with the voltage on this crystal when the rubber sample is removed. This method at low frequency is open to a number of serious objections but the aim of these experiments has been to find whether different rubbers are good or bad absorbers of ultrasonic energy. The main errors are (a) the transmitted waves are not plane and waves passing through the rubber diverge or converge, depending upon whether the velocity of sound in the rubber is greater or less than that in water; (b) air bubbles are trapped in the rubber sample, and cause scatter; (c) reflexions occur at rubber interfaces if the acoustic resistivity of the rubber differs greatly from that of water; (d) surface air must be eliminated from the sample; (e) standing waves are set up between the crystals and sample.

The absorption in the water is assumed negligible. Error (b) is eliminated in most cases by high pressure moulding. Error (d) is eliminated by soaking samples in water for long periods. Errors (a) and (c) are very small for rubbers with an acoustic resistivity similar to water. Error (e): at 50 kc/s frequency the standing waves are not clearly defined because of the wide beam spread. Fortunately at the higher frequency, the absorption increases and reduces the standing wave errors.

The absorption (a) is expressed in db/cm using the formula:—

$$a = 20 \log_{10}(V_2/V_1).$$

$V_2$  = amplified voltage from receiver before solid inserted.

$V_1$  = amplified voltage from receiver after solid inserted.

Table 2. Absorption (a) of ultrasonic waves in rubber

		Temperature 17° C.		
	Ratio by wt. of rubber to filler	a db/cm 50 kc/s frequency	a db/cm 150 kc/s	a mdb/c 350 kc/s
<i>Natural rubber base.</i>				
(1) Uncured pure rubber	—	<0.1	0.776	1.28
(2) Rubber/carbon	100/47	<0.1	0.1	0.78
(3) Rubber/carbon	100/140	<0.1	1.24	3.18
(4) Rubber/iron oxide	100/54	<0.1	<0.1	1.02
<i>Synthetic rubber base.</i>				
(5) Butyl	100/0	0.4	3.2	10.0
(6) Butyl/carbon	100/40	1.0	3.2	11.5
(7) Butyl/carbon	100/70	0.96	4.0	11.0
(8) Hycar O.R./carbon	100/4	0.40	2.9	5.1
(9) Hycar O.R./carbon	100/100	0.90	3.2	5.6
<i>Plastic.</i>				
(10) Polythene	—	1.2	1.2	2.4
(11) Polyvinylchloride 30% by weight dioctyl phthalate.	—	0.30	0.5	0.30

The figures for a number of rubbers are given in Table 2. The interesting facts of these results is that



natural rubbers transmit ultrasonic waves of low amplitude well, but the synthetic rubbers, particularly butyl and hycar rubber, are good absorbers, particularly at frequency 350 kc/s. The variation of absorption with frequency for a number of different rubbers at room temperature is shown in Fig. 5. It was interesting to find that the red rubbers loaded with iron oxide gave readings greater than when there was only water between

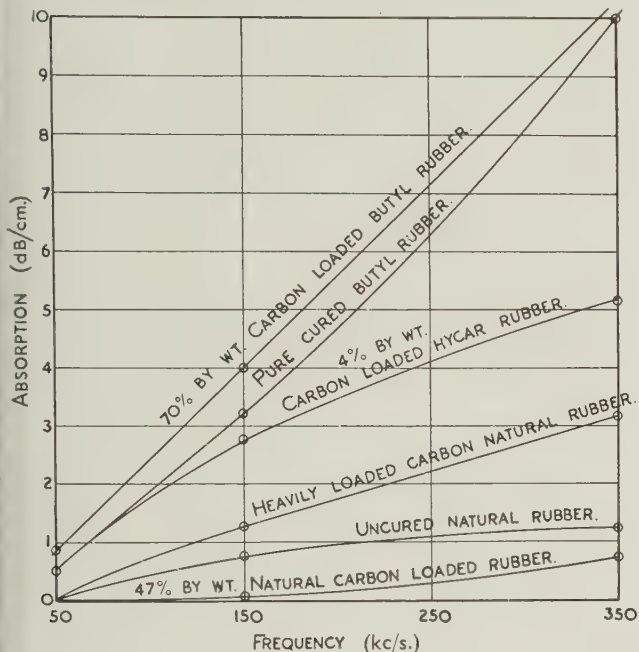


Fig. 5. Variation of ultrasonic absorption with frequency for rubbers taken from Table 2. Temperature 17°C.

the crystals. This was because the focusing action of these rubbers is greater than the absorption, and indicates one of the limitations of this method.

#### $\rho c$ RUBBER AND TRANSMISSION PLATES

There is a need in low frequency ultrasonic work for solids which will transmit ultrasonic waves at solid/liquid interfaces without reflexions and with low absorption. Rubber can easily perform this function and reference has been made<sup>(2)</sup> to  $\rho c$  or  $sc$  rubber, where  $c$  is the ultrasonic velocity of the wave in rubber and  $\rho$  or  $s$  is the specific gravity. The term  $\rho c$  rubber has been applied to rubbers with the same acoustic resistivity as the liquid.

A common application of  $\rho c$  rubber is to stick it to a piezo-electric crystal which is to transmit into a conducting liquid. The rubber gives maximum transmission and prevents the electrodes from short circuiting when a conducting liquid such as water is used. A typical crystal holder for this purpose is shown in Fig. 6. The essential feature of this crystal holder is a rubber bung stuck to the crystal with a good rubber adhesive. The rubber bung is pressed into a brass cylinder to prevent the ingress of moisture. It is also found that surrounding the rest of the crystal with foamed latex such as

Dunlopillo, serves the double purpose of giving the crystal an air backing to ensure maximum forward radiation and keeping the crystal gently but firmly clamped. Normally X-cut crystals are clamped at their mid point, but we have found that this end type of clamping is satisfactory.

Rubber and plastics make ideal transmission plates, when it is required to transfer ultrasonic energy from

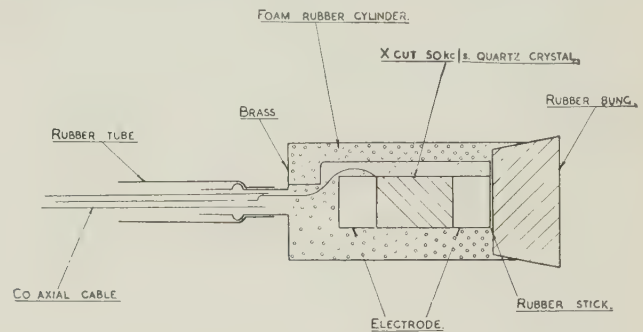


Fig. 6. Typical crystal holder

one liquid to another. It is easy to choose a rubber or plastic which is not soluble or attacked by either liquid. It is essential that the transmission plate should be kept as thin as possible, particularly at the higher ultrasonic frequencies, in order to minimize the absorption losses. For high-powered ultrasonic work using oil as one liquid, plasticized 0.003 to 0.004 in p.v.c. sheeting, and polythene are preferred. The latter has a lower reflexion coefficient at the oil-plastic boundary, but the former is easier to handle in thin sheets. It is essential to obtain p.v.c. sheet free of any minute air bubbles or impurities; otherwise after a very short time the sheet will crack. Some solvents, e.g. ketones, slowly dissolve the p.v.c. so polythene, which is very insoluble in solvents, is used. Commercial polythene beakers have proved satisfactory for transmitting ultrasonic energy. The reflexion coefficient at an oil/thick p.v.c. interface is 56% (acoustic resistivity p.v.c.  $3.45 \times 10^5$  from Table 1), but at 1 Mc/s frequency, and using 0.003 in thick p.v.c., the reflexion coefficient is 5% as calculated by Rayleigh's formula.<sup>(3)</sup>

For some ultrasonic work, particularly emulsion formation, glass vessels are preferred because certain emulsions coagulate, if irradiated for too long a time using the full available power obtained with plastic transmission plates.

#### ACOUSTIC LENSES

Acoustic lenses have been discussed by Ernst.<sup>(3)</sup> Rubbers which have an acoustic refractive index compared with water of between 1.3 and 0.7 have been used as lenses, although their focal length tends to be too long for most practical purposes. However, rubber pads placed on the ends of piezo-electric crystals alter the lobal pattern of the crystal. At low ultrasonic

frequencies the beam from a 2.5 cm diameter crystal sending out waves into water is very divergent, the semi-angle of the primary beam is about  $60^\circ$ . Plano-convex lens made from rubber, loaded with iron oxide, reduce the semi-angle of the beam, the amount depending on the velocity of sound in the rubber. From the values of velocity quoted in Fig. 3 it is possible to estimate roughly the effect such rubbers have on the lobal pattern.

It is useful to have a rubber or plastic which can easily be compounded to have a definite velocity of sound or acoustic resistivity, although it is not always possible to satisfy both conditions. Double convex lenses of long focal length, made of natural rubber loaded with iron oxide, have been used to focus ultrasonic waves. High powered experiments have been carried out at frequencies of 300, 650, and 1 000 kc/s using a 100 W ultrasonic generator supplied by The General Electric Co. Ltd. These waves are best focused by using a watch glass<sup>4</sup> but we have attempted to focus such waves with moderate success using plastic or rubber lenses. The type of lens used was a double concave moulded product with radius of curvature 1.5 cm. Identical lenses were moulded in polystyrene, polyvinyl chloride, Hycar O.R., and machined from Perspex. The polystyrene lens had the shortest focal length. Normally, the height of the oil fountain from a 100 W source is 2 cm, but inserting a polystyrene lens causes a fountain 50 cm high. After a few seconds, minute gas bubbles form under the lens and reduce its efficiency; also the lens heats up very

rapidly, and the velocity of the waves through the polystyrene decreases and alters the focal length. Both polystyrene and Perspex get sufficiently hot to cause the lenses to crack when using high ultrasonic powers, but at low powers these lenses are satisfactory.

#### CALORIMETRY

A rough method for measuring the power of ultrasonic sources, which errs on the lower side, is to use a block of polythene or rubber which is placed close to the transmitting quartz. Both these materials reflect very little energy, and if the block is made thick enough it absorbs all the ultrasonic energy. The temperature rise in the polythene can be measured at different points in the block by thermocouples and the available ultrasonic power can be calculated.

#### ACKNOWLEDGMENTS

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## The Use of Platinum Thermocouples in Vacuo at High Temperatures

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[Paper received 23 June, 1950]

In the course of thermal analysis on cobalt base alloys in vacuo with platinum-platinum 13% rhodium thermocouples it has been found that the e.m.f. developed at any temperature gradually falls. The principal cause of this is believed to be the transfer of rhodium to the platinum wire via the vapour phase. It has been shown that this effect can be reduced considerably by the use of a platinum 1% rhodium wire in place of the platinum wire.

In the course of thermal analyses in vacuo on cobalt base alloys at temperatures up to  $1\,625^\circ\text{C}$ , it has been found that the calibration of a Pt/Pt13Rh thermocouple has changed during use. Fig. 1 gives some results obtained with cobalt metal in successive calibrations after various times of immersion in the molten metal. Between cooling curves the temperature was raised to  $1\,500$ – $1\,520^\circ\text{C}$ . The value taken for time 0 is that calculated from the e.m.f. measured at the melting point of silver, prior to this melt. The wires were 0.020 in. diameter and fitted into recrystallized alumina refractories. The sheath was 5 mm outside diameter with 0.7 mm thick walls, and could be lowered a distance of 3 in into the melt without raising the pressure above the usual value ( $\frac{1}{2}$ – $5\mu$ ).

#### INVESTIGATION OF THE CAUSE OF THE DECREMENT

A used couple was analysed spectrographically with the following results:

Pt 13% Rh wire	Cu*, Si*, Mg*, Ag, Au, Be, Co, Fe, Mn, Mo, Ni with a suspicion of tin.
Pt wire	As above plus rhodium.

\* More than in the unused wire.

There was no rhodium concentration gradient between a point near the junction in the platinum wire and a point 18 mm away, although the other impurities were probably in greater concentration 18 mm from the junction. Spectrographic analysis of the refractories gave:—

Insulator (double bore)	Ag, Cu, Si
Sheath	Ag, Cu, Si, Fe, Mg, Ni



After use cobalt, chromium, molybdenum and lead were present on the sheath. Melts containing high purity cobalt, nickel, chromium and molybdenum had been made, so that the presence of these impurities was not surprising. It may be significant that chromium was absent from the wire after use.

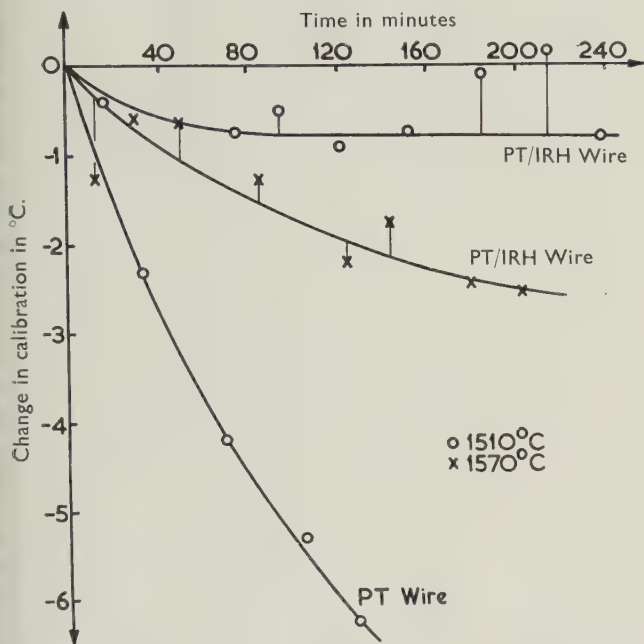


Fig. 1. The change of calibration of Pt/Pt13Rh and Pt1Rh/Pt13Rh couples after various times of immersion in molten cobalt at about 1 510° C. The change of calibration of the Pt1Rh/Pt13Rh couple in molten palladium at about 1 570° C is also shown.

These impurities were present in sufficient quantities to be the probable cause of the decreased e.m.f. given by the couple. A distinction may be drawn between the rhodium which only contaminates the platinum wire and the other impurities which are present in both wires.

#### DISTRIBUTION OF THE CONTAMINATION

(a) *Lengthwise.* This was investigated by two methods. In the first, new thermocouple junctions were made at various points equidistant along each wire from the junction, and in the second, the thermocouple was immersed to a gradually increasing depth in a bath of pure silver. Neither gives an indication of the exact distribution along the wire, because the e.m.f. is influenced by composition variations along the wires, but both gave somewhat similar results. Fig. 2 gives the e.m.f. at the freezing point of silver against the distance of the junction from the original junction for a constant depth of immersion of 18–20 mm. This was chosen because it is about the same as that used in the cooling curves on 50 g melts of cobalt base alloys. Two steps can be seen: the one at a distance of 2–3 mm away from the junction is believed to be due to the protection afforded by the insulators, and the second at 20 mm

corresponds to the depth of immersion of the couple in the molten cobalt-base alloys. The latter point will also correspond to the beginning of a rapid fall in temperature.

(b) *Diametral.* The couple was heated in a current of air for 4 h at 1 070° C followed by 2 h at 1 400–1 450° C, but ten calibrations at the silver point during this period gave e.m.f.'s between 9.882 to 9.912 mV, of which eight were within the range 9.882 to 9.887 mV. Two effects might have occurred: either the impurities

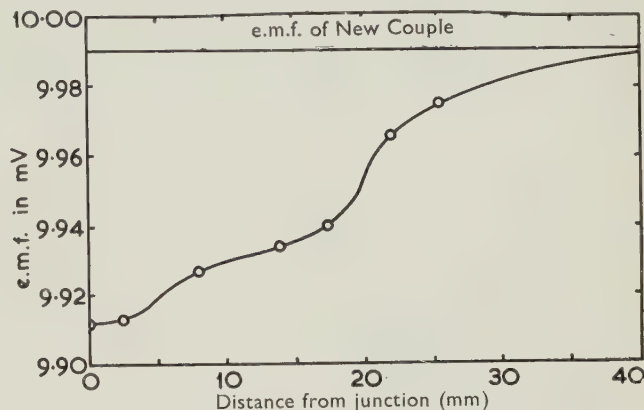


Fig. 2. The e.m.f. at the freezing point of silver for junctions formed at various distances along a contaminated couple.

would diffuse into the platinum and alloy wire or they would be oxidized away to give a certain amount of restoration of the e.m.f.<sup>(1)</sup> It is thought to be unlikely that these two effects would balance each other out over this period, if they were operative, and so the experiment was discontinued.

#### DISCUSSION OF THE CAUSE OF THE DECREMENT

The impurities present in both wires may, in some cases, exert opposite effects on the e.m.f. so that their total effect will not be as pronounced as it might otherwise be. But although no conclusions can be drawn with regard to these impurities, Stauss's results<sup>(2)</sup> show that a very small percentage of rhodium in platinum causes a large change in the e.m.f. As little as 0.004% causes the e.m.f. at 1 200° C to fall by 70  $\mu$ V.

McQuillan<sup>(3)</sup> has discussed the use of these couples in various atmospheres and concludes that diffusion of rhodium from the bead along the wire is an appreciable effect. Although no data is available on the rate of the diffusion, the lack of a concentration gradient between a point 2 mm from the junction a point 18 mm away suggests that this cannot be the mechanism of transfer in vacuo. The volatility of platinum is given as twice that of rhodium at 1 300° C.<sup>(4)</sup> Consequently, the composition of the alloy wire will tend to become enriched in rhodium, although the effect will be very small because the rhodium content of the vapour around it will be reduced by dilution with the vapour from the pure platinum wire. On the other hand, this diffusion of rhodium vapour into the proximity of the platinum

wire will upset the equilibrium here which will cause the platinum wire to take up rhodium. The rate at which rhodium is taken up by the platinum wire will depend on the composition of the vapour and the amount of rhodium already deposited on the surface. The composition of the surface of the wire will also depend on the rates of diffusion inwards and the rate of evaporation from the surface. However, when the wire contains no rhodium the rate of uptake will be a maximum. The change of e.m.f. is also a maximum being at the rate of 15–20 mV for a change of rhodium content of 1%. Fig. 3 shows that at 13% rhodium the rate has

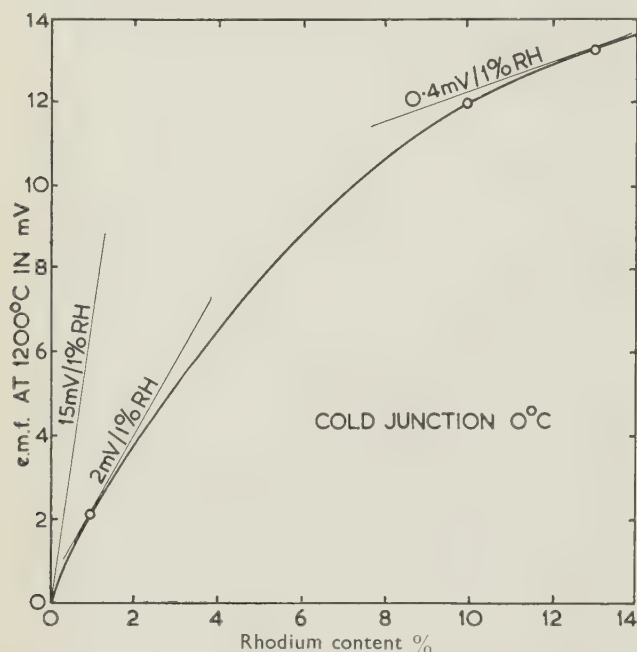


Fig. 3. The e.m.f. of Pt-Rh alloys against pure platinum at 1200°C showing the effect of small composition changes on the e.m.f. at various compositions.

fallen to 0.4 mV per 1% rhodium. Not only is the composition change of the alloy wire due to evaporation very much smaller, but the effect on the e.m.f. of a small composition change is also much smaller, so that it may be neglected in comparison with the change in the e.m.f. occurring in the platinum limb of the thermocouple.

#### THE Pt1Rh V. Pt13Rh COUPLE

The platinum wire is most susceptible to change, so that a trial has been made of a Pt1Rh v. Pt13Rh couple. The rate of change of the e.m.f. at 1% rhodium is only 2 mV/1% Rh, whilst the rate of uptake of rhodium will be reduced considerably due to the presence of the 1% rhodium already there. It has been found that immersion for 4 h in molten cobalt at an average temperature of 1510°C caused a fall in the calibration amounting to 0.8°C, whereas after 2 h the standard Pt/Pt13Rh couple had changed its calibration by 6°C (see Fig. 1). A 3½ h immersion in molten palladium at an average temperature of 1570°C caused a further fall

of 2½°C. In both cases the pressure was kept within the range 3 to ½ microns mercury.

Three calibration points have been obtained with this couple:—

Freezing point of silver . . . 960.8°C–8.206 mV

Freezing point of cobalt . . . 1493°C–14.628 mV

Freezing point of palladium . . . 1552°C–15.341 mV

The melting point of cobalt has been taken to be 1493°C Celsius instead of the value of 1492°C given in the recent paper,<sup>(5)</sup> since this was based on the values suggested in 1927 and 1939. A calibration curve has been fitted to these points with a similar shape to those of the standard couples, and this shows several interesting characteristics. The e.m.f.'s developed at low temperatures are lower, reducing cold-junction correction effects, but above about 1000°C the thermo-electric power becomes greater than that of the Pt/Pt10Rh couple.

The couple has been used extensively for melts and thermal analyses of cobalt base metals with good results. Twelve melts which were made with the same couple show that the change in calibration averages between ¼ and ½°C per run, at the melting point of nickel.

#### DISCUSSION OF RESULTS

There appears to be more than one reason why the replacement of the platinum-1% rhodium alloy should lead to great stability. The decrease in the rate of rhodium uptake by the platinum-rich wire is additional to the decreased effect of a small increase in the rhodium content on the e.m.f. If these two reasons alone are operative, then the couple would only show this marked improvement in vacuo, but there is reason to believe that the platinum wire is more susceptible to many impurities than the alloy wire, so that some improvement may well be maintained in air or in a protective gas. A search of the literature has revealed that Hanson found a Pt5Rh/Pt20Rh couple to be more resistant to contamination than the standard couple when working in nitrogen.<sup>(6)</sup> If this is the case, then the superiority may extend over a wider field.

#### ACKNOWLEDGMENTS

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# Static Charges on Dielectrics

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The author has previously shown that the permanent "homo" charge (of the same sign as the electrode) on electrets arise from a transfer of charge from electrode to dielectric surface, often by discharges. Using the method of the demountable capacitor this discharge process of charge transfer has been investigated on polystyrene. By assuming that breakdown occurs at a given inception voltage, persists until the voltage drops to an extinction voltage, and affects the whole area of the electrode and dielectric surface, a quantitative explanation has been given of the variation of the charges on the electrode and on the dielectric surface when the voltage is varied in steps, in cycles, continuously and alternating. The explanations agree with the current theories of internal and external discharges. The extinction voltage is found to be not much less than the inception voltage, as opposed to the low extinction voltages often encountered in internal discharges.

## 1. INTRODUCTION

In a recent investigation of the behaviour of the carnauba wax electret it has been shown that the electret carries two types of charge, the heterocharge and the homocharge. The first, which is of opposite sign to that of the electrode, is associated with dielectric absorption, and the second is of the same sign as the electrode and is associated with transfer of charge between the dielectric and the electrode.<sup>(1)</sup> Dielectric absorption has already been the subject of numerous studies but the transfer of charge to a dielectric surface and related problems are still insufficiently known. The present paper relates the results of a study of the formation and behaviour of real charges on dielectric surfaces. The purpose of the paper is to obtain information about the nature and distribution of these charges.

It has been already stated in the earlier paper that the charging of the dielectric surface is due to discharges which occur in air-gaps between the dielectric and the electrode. Similar phenomena are observed in voids which exist inside and at the surface of high voltage insulation, as has been shown by Whitehead and others.<sup>(2)</sup> Thus the results of this paper will also give information about the breakdown process in these voids.

The method of investigation was that of the demountable capacitor already successfully used in the investigation of the electret. The dielectric with which all measurements were made was polystyrene. This is an excellent insulator, and practically free from dielectric absorption. This and other effects possibly associated with it can therefore be disregarded here.

The demountable capacitor has one electrode which, while normally resting on the surface of the dielectric, can be lifted by means of an electro-magnet. In Fig. 1 the sample is clamped to a fixed bottom electrode by means of a guard ring (always earthed). The electrode assembly is mounted inside a heater and the entire system put into a desiccator. A detailed description has already been given in the earlier paper and the system used in the present work differed from this only in minor

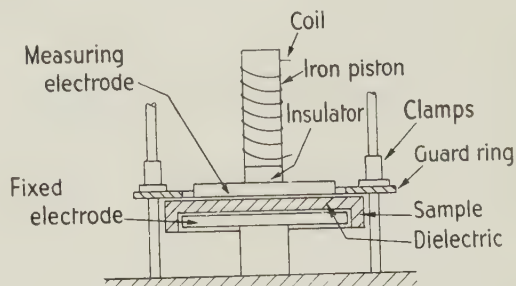


Fig. 1. Electrode assembly

features.<sup>(1)</sup> The polystyrene samples referred to herein had a thickness of 0.104 cm. The dielectric constant was  $\epsilon = 2.6$  and with an electrode area of 44 cm<sup>2</sup> the capacitance was  $C = 9.6 \times 10^{-11}$  Farad.

In the measuring circuit, shown in Fig. 2, the bottom electrode can be earthed or connected to the high voltage supply. The movable top electrode can be earthed or connected to a Lindemann electrometer and a high grade mica capacitor. The capacitance was always high enough so that the potential of the measuring electrode did not exceed 2 volts which means that the electrode for all practical purposes remained at earth potential. The readings were taken by means of the null method of Townsend in which a balancing charge was induced from a variable potentiometer.

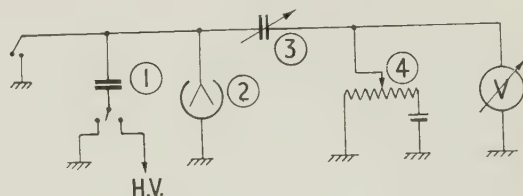


Fig. 2. Measuring circuit

- (1) Demountable capacitor.
- (2) Lindemann electrometer.
- (3) Variable mica capacitor.
- (4) Backing potentiometer.

The measuring electrode in its normal position rests on the surface of the dielectric and is earthed. If no voltage is applied and the dielectric is uncharged, e.g.

\* At present at the British Electrical and Allied Industries Research Association, Greenford, Middlesex.

does not carry any sort of surface or space charge, the measuring electrode too will have no electric charge. If voltage is applied and/or the dielectric is charged, the measuring electrode will also carry a charge. For the purpose of measuring this charge  $q$ , the plate is disconnected from earth and lifted. This procedure will charge up the electrometer-capacitor system of given capacitance  $C_0$  to a voltage  $V = q/C_0$ . Measurement of  $V$  therefore gives  $q$ . Immediately after measurement, the plate is lowered and earthed. This measurement can be made while the other electrode is earthed or while it is connected to the high voltage source.<sup>(3)</sup>

## 2. DISCHARGE THEORY

**2.1 Equivalent System.**—Even with the measuring electrode resting on the dielectric, there will always be an interface between the dielectric and the plate, the electric properties of which will approximate those of air. When the electrode is lifted the thickness of the interface will increase. The demountable capacitor therefore can be treated as a two-layer capacitor, having an air gap of dielectric constant  $\epsilon_1 = 1$  and thickness  $d$ , and a dielectric layer of thickness  $D$  and dielectric constant  $\epsilon_2 = \epsilon$ . Let  $A$  be the area of the plate in  $\text{cm}^2$ ,  $V$  the applied voltage, and suppose that the dielectric carries a real surface charge of density  $\sigma$ . The field  $E$  in the interface is then given by

$$E = \frac{\epsilon V/D - k\sigma}{1 + \epsilon d/D}, \quad V/\text{cm} \quad (1)$$

where  $k = 1.13 \times 10^{13} \text{ Vcm/coul}$ . Introducing an equivalent net charge density  $m$  of the dielectric, one gets

$$km = -\epsilon V/d + k\sigma \text{ coul/cm}^2 \quad (2)$$

The field can also be expressed in terms of the charge  $q$  of the electrode as

$$E = kq/A, \quad V/\text{cm} \quad (3a)$$

When  $d = 0$ , it follows that

$$q/A = -m \text{ coul/cm}^2 \quad (3b)$$

Thus  $m$  is equal and opposite to the charge density on an electrode of a capacitor energized by a voltage  $V$ , that is in perfect contact with a dielectric surface carrying a total (real) surface charge of density  $\sigma$ . In practice equation (3b) will hold, provided the surfaces of the dielectric and the electrode are carefully ground and polished so that  $\epsilon d/D \ll 1$ . When in particular  $V = 0$ , then  $q = -A\sigma$ . When in such conditions the electrode is lifted,  $q$  gives directly the total surface charge of the dielectric.

The equations, derived for an interface of fixed thickness, of course, remain valid when the thickness of the air-gap changes during the movement of the electrode. Thus equation (1) shows how the field in the air-gap varies during the movement of the electrode.

**2.2 Maximum Measurable Charge Density.**—Equation (3b), on which the measurement of the charges is based, depends on the condition that the electrode is in perfect contact with the dielectric. Here  $q$  is the charge of the electrode before it is lifted. However, the quantity of electricity carried by the electrode *after* lifting is equal to that carried *before* the lifting only so long as no discharges occur *during* the lifting. But when the equivalent net charge density  $m$  is sufficiently high, discharges will occur. In a highly energized capacitor these discharges will charge up the dielectric; in a capacitor which, after having been energized, is short-circuited, they will discharge the dielectric surface. The net result is that they will keep  $q$  below a maximum value.

In Fig. 3 curve (1) gives the breakdown strength of air at N.T.P. between flat electrodes as a function of the distance between the electrodes; curve (2) gives the field (as function of the thickness of the air-gap) produced by a dielectric sample, with the properties and dimensions of that used in the present experiments, with a charge of value

$$m_0 = 1 \times 10^{-8} \text{ coul/cm}^2 \quad (4a)$$

calculated from equation 1.

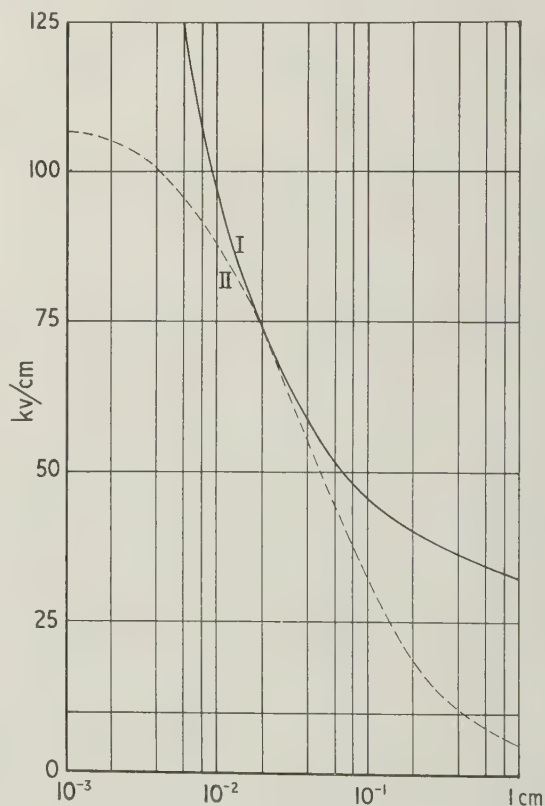


Fig. 3. Determination of breakdown threshold and maximum obtainable surface charge

- (1) Breakdown field for air.
- (2) Field of charged dielectric.



Curve (2), although normally below curve (1), touches it at a point corresponding to

$$E_0 \simeq 73\,000 \text{ V/cm} \quad (4b)$$

$$d_0 \simeq 2 \times 10^{-2} \text{ cm} \quad (4c)$$

So long as the charges are smaller than  $m_0$ , the varying field in the air-gap during the movement of the electrode will always be smaller than the breakdown strength and no discharge will occur. When, however, the charge of the dielectric is higher than  $m_0$ , then during the movement of the electrode a position will be reached where the field in the interface reaches the breakdown strength and breakdown will occur. This will reduce the charge to at least  $m_0$ . It is here supposed that the duration of the discharge is sufficiently small so that in spite of the movement of the electrode the system can always be considered as being in equilibrium.

It follows that in the present experiments  $1 \times 10^{-8}$  coul/cm<sup>2</sup> should be the maximum obtainable charge density. Higher charges could be obtained if thinner samples were used.

The operational characteristics of the demountable capacitor are seen to be the same as that of a capacitor with fixed electrodes having an air-gap of thickness  $d_0$  between the dielectric and one of the electrodes.

The foregoing discussion has shown how breakdown keeps the measurable charge density below  $m_0$ . Further, it will readily be seen that charging of the dielectric is achieved when the energized electrode is lowered toward the dielectric.

**2.3 Charge-Voltage Characteristics.**—The nature of the phenomena to be expected will depend on the way the capacitor is operated. There are two main methods, one suitable for the measurement of the charge of the short-circuited dielectric, the other for the measurement of the charge of the energized capacitor. The *first method* proceeds in two steps.

(a) A voltage  $V$  is applied to the capacitor, the movable electrode is lifted, the charge  $q_1$  measured, and the electrode lowered again.

(b) The voltage is disconnected, the system temporarily short-circuited, the electrode lifted again and the charge  $q_2$  is measured. This procedure is repeated with different voltages.  $q_1$  will be the charge residing on the electrode of the energized capacitor,  $-q_2$  will be the charge on the dielectric.

Starting with an uncharged dielectric,  $q_1$  will at first increase with  $V$  as

$$\left. \begin{aligned} q_1 &= CV \\ \sigma &= 0 \end{aligned} \right\} \quad V < V_0 \quad (5a)$$

Eventually, however, a threshold voltage  $V_0$  is reached sufficient to produce breakdown in the air-gap during the lifting of the electrode. If, to a first approximation, equality of extinction and inception voltage is assumed, then, for values  $V > V_0$ ,  $q_1$  will remain constant, while the charge of the dielectric will start to rise with the same

slope as did previously the charge of the electrode. (Fig. 4.)

Therefore

$$\left. \begin{aligned} q_1 &= Am_0 \\ A\sigma &= -q_2 = C(V - V_0) \end{aligned} \right\} \quad V_0 < V < 2V_0 \quad (5b)$$

When, however,  $V$  has attained twice the threshold value  $V_0$ ,  $-q_2$  will have reached the saturation value  $Am_0$ ; the dielectric is now so highly charged that it will produce a back-discharge during stage (b) of the experiment and

$$q_1 = -q_2 = Am_0 \quad V > 2V_0 \quad (5c)$$

The threshold voltage  $V_0$  is that voltage which according to equation (1) produces with an uncharged dielectric ( $\sigma = 0$ ) the field  $E_0$  for a gap of width  $d_0$ . Thus

$$V_0 = (1 + \epsilon d_0/D) DE_0/\epsilon \quad (6)$$

With the data of the present experiments one gets  $V_0 = 4\,400 \text{ V}$ .

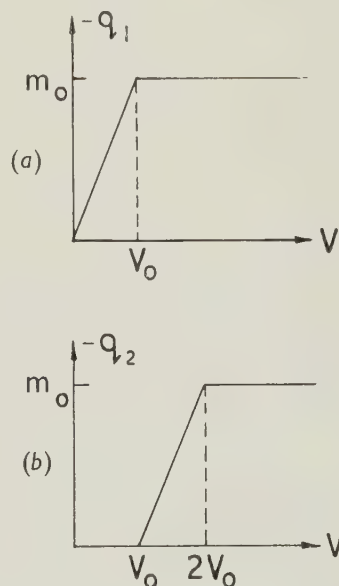


Fig. 4. Charges of the electrode (a) and of the dielectric (b) as function of applied voltage

Once the dielectric has been charged up to a certain charge  $A\sigma = -q'_2$  by application of a voltage  $V'$ , a field in the contrary direction may be applied. Since now the external field and the field of the polarized dielectric act in the same sense, a breakdown field in the air-gap will be attained at voltages smaller than  $-V_0$ . Equation (1) applies again, but  $\sigma$ , instead of being 0 as before, now has the value  $\sigma = C(V' - V_0)/A$  obtained from equation (5b). Observing that  $C = \epsilon A/kD$  one gets

$$E_0 = \frac{-\epsilon V^*/D - \epsilon(V' - V_0)/D}{1 + \epsilon d_0/D} \quad (7)$$

where  $-V^*$  is the new breakdown threshold after

inversion.  $E_0$  can be expressed in terms of  $V_0$  by equation (6). The final result is

$$-V^* = \begin{cases} V_0 & V' \leq V_0 \\ V_0 - (V' - V_0) & V_0 < V' < 2V_0 \\ 0 & 2V_0 \leq V' \end{cases} \quad (8)$$

After the threshold is reached the charge of the dielectric will at first decrease, go through zero, and increase with the contrary polarity as before. The slope of the curve is again proportional to  $C$ . When the applied voltage  $V$  goes through a complete cycle, from  $+V'$  to  $-V'$ , and back again to  $+V'$ , the charge ( $-q_2$ ) of the dielectric plotted as a function of  $V$  will follow a hysteresis loop. The different types of hysteresis loops corresponding to different values of  $V'$  are shown in Fig. 5a.

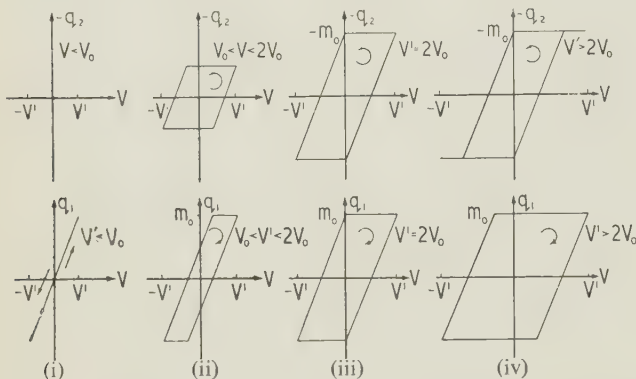


Fig. 5. Charge hysteresis. (The upper diagrams refer to the charge of the dielectric. The lower diagrams refer to the charge of the electrode of the energized capacitor.)

- (i) Low voltage. Peak voltage below breakdown threshold. No discharges occur.
- (ii) Low voltage. Peak voltage slightly above threshold. Discharges occur during part of rising cycle.
- (iii) Medium voltage. Peak voltage equal to twice threshold. Discharges occur over entire rising cycle.
- (iv) High voltage. Peak voltage exceeds twice threshold. Discharges occur also over part of falling cycle.

The method of measurement so far described gives direct information about the behaviour of the charge on the dielectric. There is, however, the objection that in practice, when for instance a voltage surge or a.c. stress is applied to a capacitor, there is obviously no periodical short-circuiting of the system like that occurring between the two measurements in stage (b) of the experiment. In practice the charge in the dielectric will therefore build up beyond the value  $m_0$  which only represents the maximum directly measurable charge. In the region in which back-discharge occurs the *measured* charge values will differ from those which actually exist on a dielectric exposed to a continuously changing stress. Since it is impossible to measure directly the charge on the dielectric prior to the back-discharge, reliance has to be placed on the *second method* of operation, mainly observation of the charge of the energized capacitor.

A voltage  $V$  is applied, the electrode is lifted and the charge measured; after lowering the electrode the system

is not short-circuited, but a different voltage is applied and the charge measured again, and so on. When the applied voltage goes through a cycle, from  $+V'$  over  $-V'$  to  $+V'$ , hysteresis loops similar to those described before will be observed. They are easily obtained from the condition that  $q_1$  can never exceed  $m_0$ . According to the value of  $V'$  one will then get curves of the form indicated in Fig. 5b.

In practice several effects will make the hysteresis loops differ considerably from the ideal curves just described. So far it has been supposed that the field is that of a perfect plane capacitor. In reality in consequence of the edge effect and unavoidable surface irregularities the field will be enhanced in places. With increasing voltage this will then lead to local discharges below the theoretical breakdown threshold; it will also reduce the threshold for the back-discharges and therefore reduce the saturation charge. The resulting hysteresis loop is of the type given in Fig. 6a. A further factor to be taken into account is the difference between discharge inception and extinction voltages. Once a discharge is initiated at a field  $E_a$  it will go on until the field has fallen down to the smaller value  $E_b$ . As a result the charge hysteresis curves are not continuous but discontinuous ones (Fig. 6b).

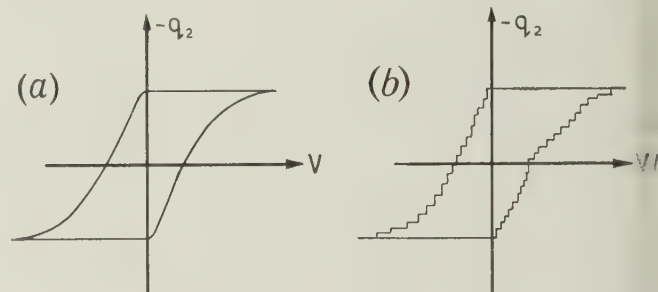


Fig. 6. Charge of the dielectric

- (a) Point discharges reduce breakdown threshold and maximum obtainable charge.
- (b) Hysteresis curve is made discontinuous by difference of discharge inception and extinction voltage.

### 3. EXPERIMENTS

**3.1 Breakdown Threshold.**—When increasing voltages are applied, then according to theory a point should eventually be reached where the charge of the electrodes should increase no further, and at the same time the dielectric should begin to acquire a surface charge. This experiment is shown in Fig. 7. Curve (2) gives the charge found on the dielectric after earthing, curve (1) the charge of the electrode obtained while voltage was applied, and curve (3) the sum of (1) and (2). The transitions between the horizontal and the inclined sections of the curves are seen not to be so sharp as predicted by the simple discharge theory, but still sharp enough to determine a definite threshold voltage: curve (2) starts rising at about 3 600 V; curve (1) reaches its maximum at 4 000 V. The breakdown threshold is therefore of the order of 3 600 V. The existence of a



charge saturation value is clearly indicated by the levelling-off of curves (2) and (3) which sets in at sufficiently high voltages, of the order of 7 000 to 8 000 V. An important point is that all three curves over a fairly wide interval have a slope of the theoretically predicted value  $C$ . This fact is evidence of a complete discharge covering the entire area between the dielectric and the electrode. The maximum obtainable charge density is  $7.5 \times 10^{-9}$  coul/cm<sup>2</sup>. The charge of the electrode after having reached a maximum value drops to values smaller than those of the charge of the dielectric. This arises from the difference between discharge inception and extinction voltage. In a breakdown process in which the dielectric is charged up, the charge of the electrode will fall *below* the value at which breakdown has set in. Therefore more than "just the necessary" amount of charge is transferred to the dielectric, the charge of which therefore will surpass that of the electrode, until at sufficiently high voltages the back-discharges (during the lifting of the earthed electrode) will affect in a similar way the charge of the dielectric. Therefore curve (2) also will sooner or later reach a maximum and then drop slightly. Apparently this occurs at still higher voltages than those reached in these experiments.

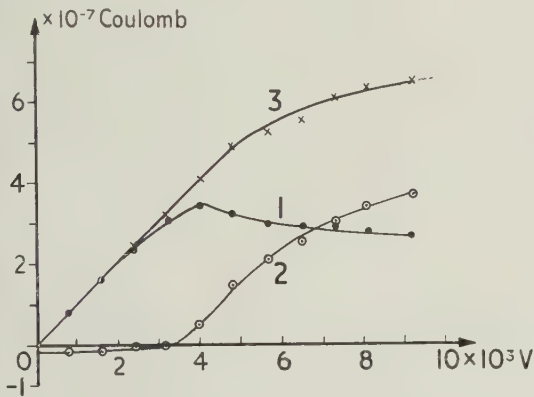


Fig. 7. Breakdown threshold and charge saturation

- (1) Charge of electrode.
- (2) Charge of dielectric.
- (3) Charge of electrode + charge of dielectric.

The experimental values for the breakdown threshold and maximum charge density coincide sufficiently well with the theoretical values. The fact that both the experimental values are smaller than the theoretical ones is attributed to reduction of the breakdown threshold by local discharges. The observed difference between inception and extinction voltages is similar to that encountered by others with external discharges. With internal discharges the extinction voltage appears sometimes to be quite low.

### 3.2 Hysteresis Loops of the Charge of the Dielectric.

The foregoing measurements were now extended, the applied voltage going through a complete cycle. Results are shown in Fig. 8 for different values of the peak

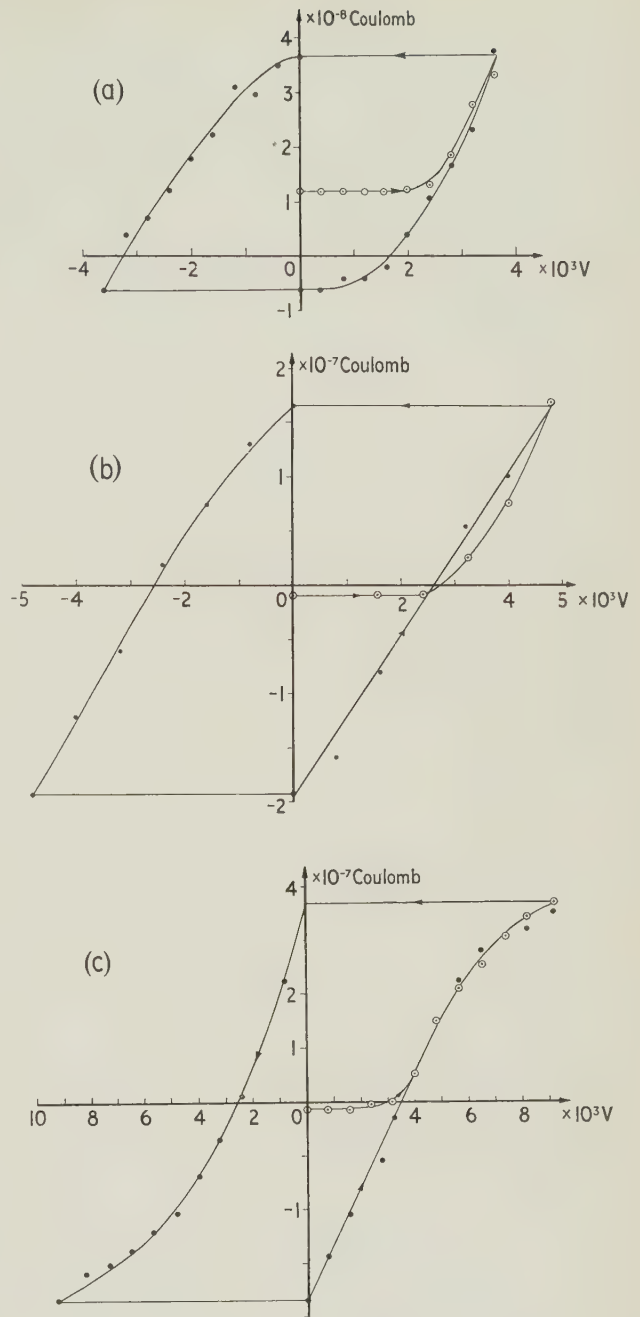


Fig. 8. Hysteresis loop of charge of dielectric  
(a) Low voltage (b) Medium voltage. (c) High voltage.

voltage  $V'$ . For Fig. 8a this was only slightly above breakdown threshold, for Fig. 8b about twice as much, and for Fig. 8c considerably higher. The characteristic hysteresis loops predicted by the theory are actually found, and these loops are seen to close remarkably well. Their shape changes according to the value of the peak voltage. At low peak voltage, reversal of the applied field does not immediately lead to a sharp bending of the charge-voltage curve; at medium and high peak voltage

this curve exhibits a sharp knee immediately after the polarity reversal; and at the high voltage the additional feature of charge saturation appears. In all three cases the inclination of the rising and of the falling parts of the curves is the same and equal to the theoretical value  $C$ . From this fact it is concluded again that the discharges in the air-gap in general cover the entire area between dielectric and electrode.

**3.3 Hysteresis Loops of the Charge of the Electrode of the Energized Capacitor.**—Next the charge of the electrode was measured as a function of voltage with the high voltage permanently applied. Measurements were

again taken over the full cycle, when the applied voltage was increased in steps up to the maximum value  $V'$ , reduced in the same steps to  $-V'$ , and then back to  $V'$ . Results are shown in Fig. 9 for rather low, medium and high peak voltage  $V'$ . Again the characteristic hysteresis effect is found. The height of the curves, which is given by the maximum charge density  $m_0$ , remains practically the same, but the width of the loops increases with increasing peak voltage. According to the theoretical exposition, discharges occur over the region where the curves are horizontal. The increase of the horizontal parts therefore shows how with increasing peak voltage the zone of discharges extends. In the low voltage case (Fig. 9a) this zone is barely distinguishable; with a medium voltage (Fig. 9b) it already extends from positive to negative voltages, and with high voltages (Fig. 9c) it extends over the greater part of the cycle. The points along the horizontal parts of the curves apparently show strong dispersion. This, however, is not fortuitous but caused by the difference between extinction and inception voltage; and the description of these parts as "horizontal" ones is to some extent an over-simplification. Considering for instance the case of rising voltages, suppose that a discharge has just occurred at the end of a voltage step; then after the discharge the field in the air-gap will have decreased below breakdown threshold. When the drop has been great enough, the air-gap field will not be restored to the breakdown value after the next, and perhaps not even after the second following voltage step. No discharges will occur in between, and the points for the two or three measurements will lie on a straight line the slope of which is equal to the capacitance of the system, and therefore parallel to the rising and falling parts of the hysteresis loops. This is shown by the dashed curves in these figures.

**3.4 Discharge Diagrams.**—The true discontinuous structure of the hysteresis loops can only be seen when the voltage is changed in sufficiently small steps, smaller at least as the voltage drop for one discharge. Measurements are shown in Fig. 10.

Fig. 10a gives the charge of the dielectric (obtained by the first method of operation); the peak voltage was not allowed to reach a value where appreciable back-discharges would occur during the short-circuiting period. The measurement is therefore unaffected by this process and gives the charge which actually would be found on the dielectric of a two-layer capacitor exposed to continuously applied stress. This charge now is seen to rise in steps only, being perfectly constant between two successive steps. The length of one such step is a measure of the (total) voltage drop corresponding to one breakdown process; roughly a value of 500 V is found. The height of a step gives the quantity of electricity discharged in each breakdown process; here a considerable dispersion is found.

Fig. 10b gives the charge of the electrode of the energized capacitor (obtained by the second method of operation of the demountable capacitor). It now

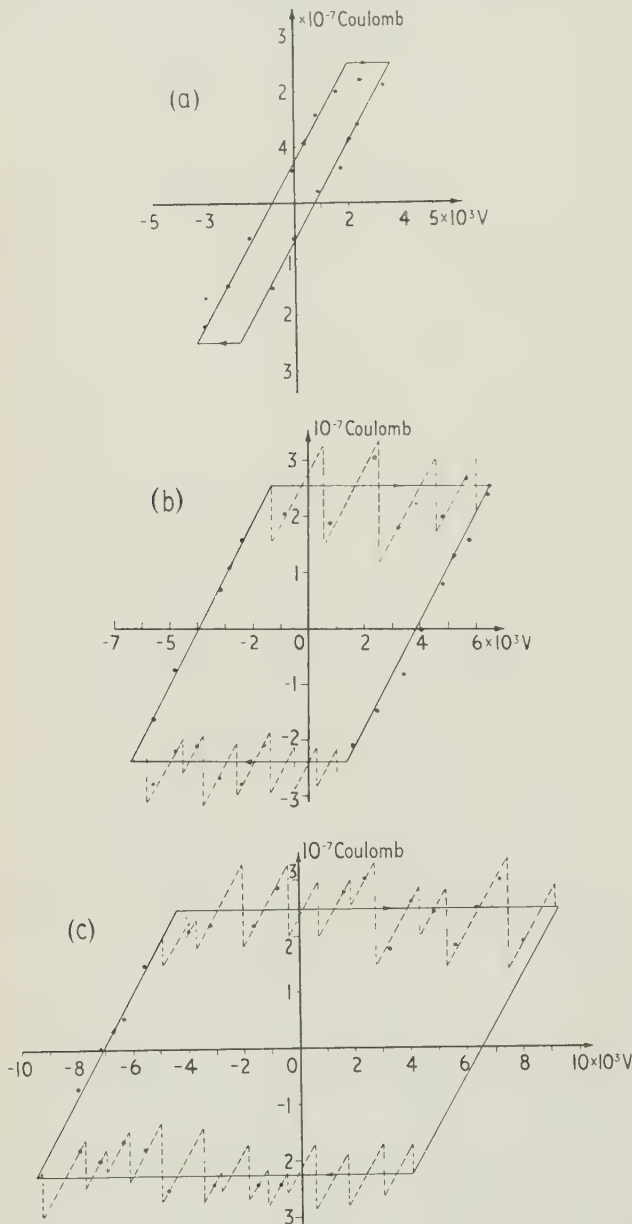


Fig. 9. Hysteresis loop of charge of electrode when capacitor is permanently energized

(a) Low voltage. (b) Medium voltage. (c) High voltage.



becomes clear that the "horizontal section" of the previous experiments really consists of a sequence of inclined straight lines which have all the same slope  $C$ , and lend to this section of the loop the aspect of the saw-tooth curve which is typical for so many discharge phenomena. The distance between two vertical lines gives again the voltage drop, and the height gives the quantity of electricity, for each discharge. Values similar to those given before are obtained. The mechanism of the phenomenon has already been explained in Section (3.3). The peak voltage for the measurement of Fig. 10b was high. Discharges occurred therefore over the greater part of the cycle.

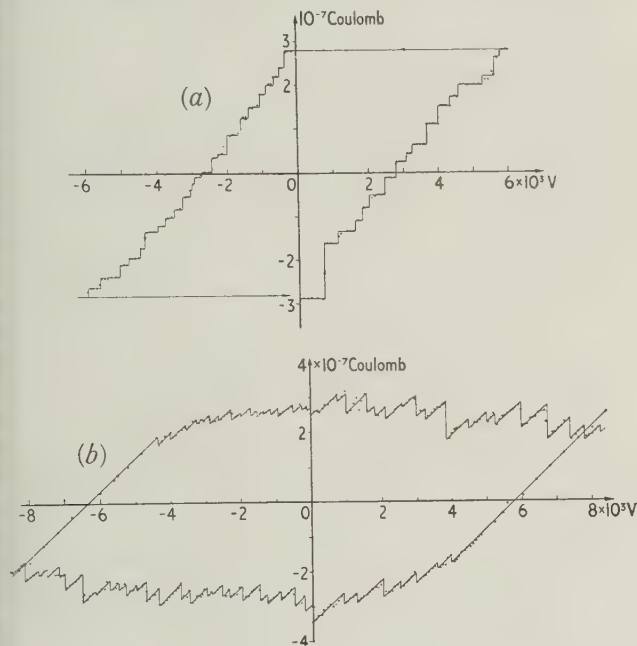


Fig. 10. Indication of discharges in the hysteresis loop  
(a) Charge of the dielectric. (b) Charge of the electrode of energized capacitor.

The foregoing measurements permit determination of the *discharge diagram*, that is a diagram which shows where and how discharges occur under alternating stress. Diagrams corresponding to the two foregoing experiments are shown in Figs. 11a and 11b, the first obtained from the measurement of the charge of the dielectric, the second from that of the charge of the electrode of the energized capacitor. The diagrams are obtained in the following way. An alternating voltage of the right peak value  $V'$  is shown. The voltages  $V_1, V_2, V_3$ , etc., where the corresponding hysteresis loops show discontinuities, are marked by small vertical lines, the length of each being proportional to the magnitude of the corresponding step. Thus each such line indicates a discharge. Their distribution shows the distribution of the discharges over the voltage swing, their length is a measure of the quantity of charge transferred in each breakdown process. With the lower voltage, breakdown

is seen to occur only during the "rising" quarter-cycles of the applied voltage; with high peak voltages, discharges are also found in "falling" quarter-cycles, before the polarity reversal of the applied voltage.

**3.5 The De-polarization Experiment.**—It can be concluded from the hysteresis loops that a dielectric once it has acquired a total surface charge can be de-polarized by the application of decreasing voltages of alternatively positive and negative sign. This has also been confirmed experimentally.

**3.6 Polarization of the Dielectric without Removal of the Electrode.**—In all foregoing experiments the dielectric was charged by lifting the electrode of the energized capacitor. However, since there is not perfect contact between the electrode and the dielectric even while the electrode is resting on the dielectric, a small amount of charging is to be expected even without lifting the electrode. This is shown in Fig. 12. Here the voltage was applied for 5 sec each time without lifting the electrode. Then the voltage was removed, the system short-circuited, and the charge of the dielectric measured as in the former experiments. The charge now is very much lower than before, reaching not even 10% of that previously obtained; in spite of this, saturation is seen to occur.

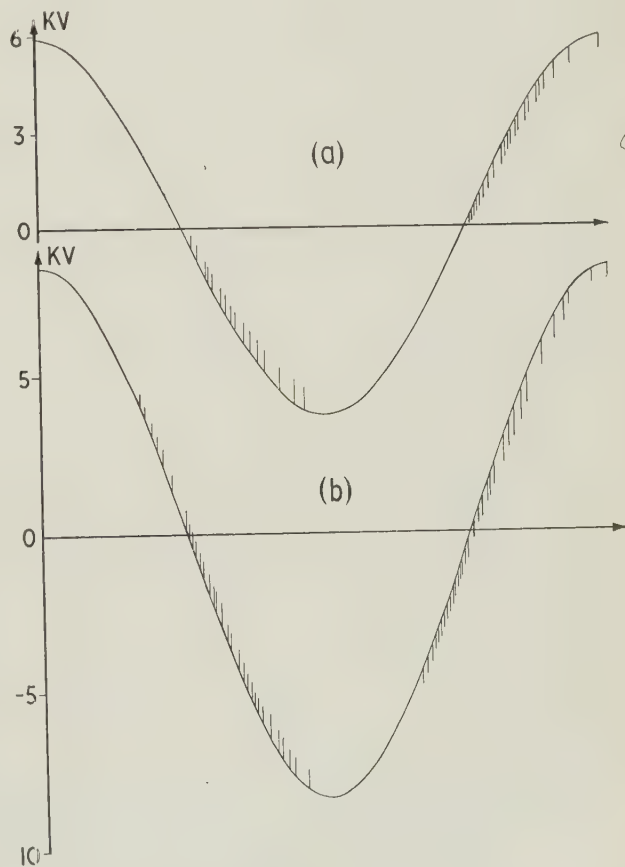


Fig. 11. Discharge diagram for alternating stress  
(a) Medium voltage. (b) High voltage.

3.7 *Two Dielectric Surfaces*.—According to the theoretical exposition, discharges between two dielectric surfaces should show the same behaviour as discharges between a metal electrode and a dielectric. Thus the measuring electrode was fitted with a polystyrene disk of 0.15 cm thickness and the charge measurements were repeated under these conditions. It was found that the general aspect of the hysteresis loop is the same as before; but the smaller field (owing to the increase of total dielectric thickness) results in a decrease of the charge and an increase in the voltage for which discharges set in.

#### 4. DISCUSSION

The agreement between the results of the experiments and the predictions of the theory is remarkably good. Theory has been based on two main assumptions: (a) the difference between the discharge inception and discharge extinction voltage is small; (b) the effects of the discharges are not localized, but extend over the entire area between dielectric and electrode. How far then do the results represent a confirmation of these theoretical premises?

Much emphasis has been placed on the evidence that the slope of the charge-voltage curves for the dielectric over a wide region equals the capacitance  $C$  of the system. To appreciate fully the significance of this point, consider the experiment referred to in Fig. 7, in which the applied voltage is increased from 0 to a maximum value, the dielectric being uncharged at the start of the experiment. There are four major regions.

(a) No discharges. The charge of the dielectric remains zero, the charge of the electrode increases as  $CV$ .

(b) Partial and/or localized discharges. The charge of the electrode still increases, but the slope of the  $q$ - $V$  curve is smaller than  $C$ . The charge of the dielectric increases also; the slope of the corresponding  $q$ - $V$  curve is also smaller than  $C$ . Here the field in the air-gap is already so high, that slight enhancement of the field due to surface irregularities leads to localized breakdown processes. In these processes the dielectric becomes charged in some places, in others it remains uncharged. The charge thus transferred to the dielectric is missing on the electrode and is found on the dielectric.

(c) Total discharges. The charge of the electrode remains constant, the charge of the dielectric increases with a slope equaling  $C$ . Here the field in the air-gap is everywhere above breakdown threshold. Therefore discharges occur over the entire area and the whole charge that otherwise should be found on the electrode is transferred to the dielectric.

(d) Back-discharges. The charge of the dielectric tends to become constant. Here the dielectric has already been charged up so strongly that back-discharges occur during the lifting of the earthed electrode.

Inspection of Fig. 7 shows clearly the existence of all four regions: there is a region below about 3 000 V,

where the charge of the dielectric remains practically unaltered and the increase of the charge of the electrode is linear. Between 3 000 and 4 000 V the charge of the dielectric increases strongly, the increase of the charge on the electrode slowing down. After 4 000 V the charge on the electrode actually decreases but the charge of the dielectric now rises with a slope equal to  $C$ . At still higher voltages this slope strongly decreases. The theory thus provides a complete and satisfactory account of the experimental results, and this in itself amply justifies the theoretical assumptions.

On the other hand, this evidence alone might not justify the exclusion of all other alternative explanations and different mechanisms could be visualized which also would explain the experimental facts. It may be assumed, for instance, that with increasing voltage the discharges still remain localized, still occur in channels and affect only small spots. To account for the fact that the total charge of the dielectric increases proportionally to the capacitance, every charged spot of the dielectric must then have a much higher charge density than for a uniform charge distribution. For such a high localized charge density to appear, the field in the area affected by the discharge channel must have fallen to a value very much lower than the breakdown threshold; this obviously would then have the result that within the area of the discharge the entire charge previously carried by the electrode is transferred to the dielectric.

Several reasons, however, seem to make this mechanism unlikely. How is it that the total quantity of charge transferred is always just the right value  $CV$ ? This seems to require an additional hypothesis. The high charge density of the charged spots of the dielectric would soon lead to back-discharges and thus not allow the total surface charge to reach the theoretical saturation value as closely as it did in the present experiments. The maximum obtainable charge density was calculated on the assumption of a uniform charge distribution. Any other distribution will yield higher field strengths and therefore lower charge values. If the  $1 \times 10^{-8}$  coul/cm<sup>2</sup> obtained for the theoretical saturation value were concentrated in charged spots, then the field at these spots would greatly exceed breakdown value. Breakdown could scarcely be prevented. In order to reduce the charge density to a "safe" level, there should also be found uncharged parts of the dielectric; but these, if present, would have repeatedly been exposed to fields much higher than breakdown value and so should retain some charge.

Consider in the light of this discussion Fig. 12, which shows charging the dielectric *without* lifting the electrodes. Here the discharges certainly can have been only partial ones, because at most parts of the surface the contact between the electrode and the dielectric must have been sufficiently good so that breakdown here can be ruled out. In this experiment the small charge of the dielectric was probably acquired in localized discharges occurring at points where the



surfaces of the dielectric or the electrode had irregularities—small points, scratches, furrows, etc. Thus all discharges being only partial ones, the slope of charge-voltage curve never exceeds even 10% of the values obtained during the previous experiments. But the surface charge of the dielectric is unevenly distributed and concentrated in spots where the charge density must be high. Therefore in spite of the low average value of the charge density, back-discharges set in soon and cause the saturation character of the curves. This shows how the maximum theoretical value  $m_0$  can never be reached when localized breakdown is the predominant feature.

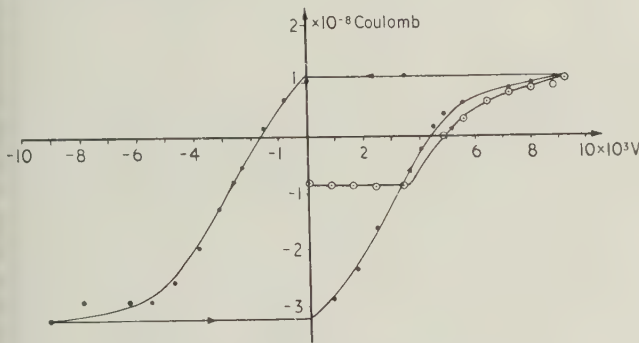


Fig. 12. Polarization of the dielectric without lifting of electrode

Similar arguments apply with regard to a theory that assumes that the discharges, although not occurring in spots, affect only limited areas of the dielectric. To explain the experimental slope of the charge curves, the charged areas must have a correspondingly higher charge density (actually if the extension of such a charged area is  $A'$ , its charge density would have to be  $A/A'$  greater than that of a dielectric charged uniformly throughout). This higher charge density would again give rise to back-discharges shortly after it has been charged and the saturation-character of the curves would have to be much more pronounced.

When a discharge occurs between two metal electrodes, an arc can be formed; the resistance of the discharge gap immediately drops to so low a value that high currents drain the electrodes and the voltage between the two electrodes drops to a value depending on the impedance of the voltage source. In consequence of this voltage drop a discharge occurring at one point between the electrodes affects the charge and potential distribution over the entire electrode area. By reducing the potential difference between the electrodes it precludes breakdown outside the discharge channel, because to start breakdown outside, the voltage would have to remain at its original value. The situation between a dielectric and a metal surface is different. The air-gap field here depends

entirely on the charge distribution of the dielectric and the potential of the electrode. When breakdown occurs at one point, the charge distribution of the dielectric at any other point remains unaffected, if the dielectric surface charge is immobile. The potential of the electrode remains unchanged because here the internal impedance of the discharge system itself is high. Therefore inter-dependence between discharges at different regions of the discharge area will not prevent breakdown from occurring elsewhere. These considerations are confirmed by the result of the measurements with two dielectric surfaces which are essentially the same as those obtained between a dielectric and a metal.

The rather low voltage drop found during the discharge seems to indicate a self-quenching action in the discharge. Possibly the high internal resistance of the system weakens the field by counter voltages and space charges. External discharges appear to differ from internal discharges in that the voltage drop in the latter may be large enough to reduce the remanent voltage to a small value.

#### ACKNOWLEDGEMENTS

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# An Electro-Magnetic Problem

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[Paper received 22 June, 1950]

A manageable form of a problem put to Oliver Heaviside in 1916 is solved. A pair of concentric spheres, of radii  $a$ ,  $b$ , and centre  $O$ , carry charges  $q$  and  $-q$ . A pole  $m$  moves along  $OX$  and gives rise to electric force in circles round  $OX$ . The pair of spheres experiences a resultant couple  $J$ , of axis  $OX$ . The pole experiences no couple. The force on the medium is  $d(V\mathbf{DB})/dt$  per unit volume, where  $\mathbf{D}$  is electric displacement and  $\mathbf{B}$  is magnetic induction. This force, like  $\mathbf{D}$ , is null except between the spheres. The total couple due to it amounts to  $-J$ .

In 1916 I sent the following electromagnetic problem to Oliver Heaviside:

A circle of radius  $a$ , with centre  $O$  and axis  $OX$ , carries an electric charge  $q$ . A magnetic pole  $m$ , moving along  $OX$  with velocity  $u$ , produces an electric force in circles round  $OX$ . When the distance of  $m$  from the arc of the circle is  $r$ , the electric force, for small  $u$ , is  $muar/r^3$ . There is, thus, a couple,  $G$ , acting on the circle, and  $G = qmuar^2/r^3$ . The pole experiences no couple. The reaction must be against the ether. The force on the ether per unit volume is  $d(V\mathbf{DB})/dt$ , and this will have a moment about  $OX$ . Here  $V\mathbf{DB}$  is the vector product of the electric displacement  $\mathbf{D}$  and the magnetic induction  $\mathbf{B}$ .

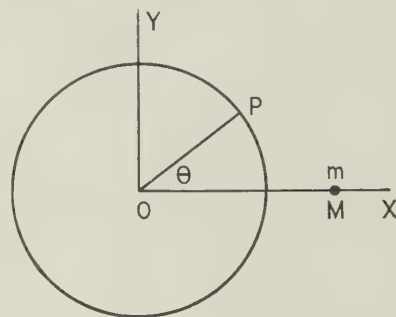
There was clearly a challenge to Heaviside to show that the couple due to  $d(V\mathbf{DB})/dt$  equals that on the charged ring. I have no record that he attacked the problem.

The force  $d(V\mathbf{DB})/dt$  occurs many times in Heaviside's treatment of the electromagnetic field, but he did not examine its consequences. The preparations for the centenary of his birth on 18 May, 1850, gave me the opportunity to see my letter to him, and led me to write this paper as a tribute to the long friendship between us.

The problem, in the form put to Heaviside, would present great mathematical difficulties. But I have found a case where (i) the couple experienced by the system of charges, and (ii) that due to the force  $d(V\mathbf{DB})/dt$  can be calculated. The two couples are found to be equal and opposite. The electrified system is formed of two spherical surfaces, of centre  $O$  and of radii  $a$ ,  $b$ , with charges  $+q$  and  $-q$ . The pole moves along an axis  $OX$ .

We consider first a sphere of radius  $R$  and centre  $O$ , with a charge  $q$ .

The pole  $m$  moves in the positive direction along an axis  $OX$  with small velocity  $u$ . At time  $t$ ,  $m$  is at  $M$  where  $OM = x$ ; thus  $u = dx/dt$ . We take  $OX$ ,  $OY$  in the plane of the paper, as shown, and  $OZ$  is towards the reader. The plane of the paper, viz.  $z = 0$ , cuts the sphere in a circle of radius  $R$ , and a point  $P$  on this circle has co-ordinates  $R \cos \theta$ ,  $R \sin \theta$ , 0, or  $\xi$ ,  $\eta$ , 0. A plane through  $P$ , with  $OX$  for normal, cuts the sphere in a circle of radius  $\eta$ , and, at any point on this circle, the electric force due to the moving pole is  $-mu\eta/r^3$ , or  $-muR \sin \theta/r^3$ , where  $r = PM$ . If  $\eta$  be positive,



the positive direction of the electric force at  $P$  is that of  $OZ$ . Here  $POX = \theta$ . We have

$$r^2 = x^2 - 2xR \cos \theta + R^2 = x^2 - 2x\xi + R^2 \quad (1)$$

For the surface density,  $\sigma$ , we have  $\sigma = q/4\pi R^2$ . The area of a ring-element of surface defined by  $\theta$  and  $\theta + d\theta$  is  $2\pi R^2 \sin \theta d\theta$ . If the moment about  $OX$  in the positive direction (i.e. from  $OY$  to  $OZ$  through  $\frac{1}{2}\pi$ ) of the force on the charge on the ring element be  $(dG/d\theta)d\theta$ , we have

$$\begin{aligned} dG/d\theta &= -(mu \sin \theta \cdot R/r^3)(q/4\pi R^2)2\pi R^2 \sin \theta \cdot R \sin \theta \\ &= -\frac{1}{2}muq(R^2/r^3) \sin^3 \theta \end{aligned} \quad (2)$$

We shall need the integral

$$S(f) = \int_0^\pi \frac{\sin^3 \theta d\theta}{(f - \cos \theta)^{\frac{3}{2}}} \quad (3)$$

where  $f = (x^2 + R^2)/2xR$ . Here  $x$  and  $R$  are sides of a triangle and are essentially positive. Then  $f > 1$ . For simplicity, we restrict ourselves to the case in which  $x > R$ . If we put  $f - \cos \theta = h$ , we have

$$\begin{aligned} S(f) &= \int_{f-1}^{f+1} \frac{1 - (f-h)^2}{h^{\frac{3}{2}}} dh \\ &= \frac{2}{3} \left[ \frac{3(f+1)(f-1) + h(6f-h)}{h^{\frac{1}{2}}} \right]_{f-1}^{f+1} \\ &= \frac{8}{3} \{ (2f-1)(f+1)^{\frac{1}{2}} - (2f+1)(f-1)^{\frac{1}{2}} \} \\ &= \frac{8}{3} \{ (2f-1)(x+R) - (2f+1)(x-R) \} (2xR)^{-\frac{1}{2}} \\ &= \frac{32}{3} (R/2x)^{\frac{3}{2}} \end{aligned} \quad (4)$$



We also have  $r^2 = 2xR(f - \cos \theta)$ . Hence

$$T(f) \equiv \int_0^\pi \frac{\sin^3 \theta d\theta}{r^3} = (2xR)^{-\frac{3}{2}} S(f) = \frac{4}{3x^3} \quad (5)$$

We now find, by (2),

$$G = -\frac{1}{2}muqR^2T(f) = -2muqR^2/3x^3 \quad (6)$$

If we have two concentric spheres, one of radius  $a$ , with charge  $q$ , and one of greater radius  $b$ , with charge  $-q$ , we see, by (6), that  $J$ , the resultant moment of the forces on the pair, is given by

$$J = 2muq(b^2 - a^2)/3x^3 \quad (7)$$

We will now find the moment about  $OX$  due to the force  $d(V\mathbf{DB})/dt$  per unit volume of the medium between the spheres of radii  $a$  and  $b$ . Since  $D = 0$  inside the smaller and outside the larger sphere, the force is null in those regions. At the point  $P$ , of co-ordinates  $R \cos \theta, R \sin \theta, 0$ , (or  $\xi, \eta, 0$ ), if  $a < R < b$ , we have

$$D_1 = \frac{q\xi}{4\pi R^3}; D_2 = \frac{q\eta}{4\pi R^3}; B_1 = -\frac{m(x - \xi)}{r^3}; B_2 = \frac{m\eta}{r^3}$$

Thus, for the  $z$ -component of  $V\mathbf{DB}$  at  $P$ , we have

$$D_1B_2 - D_2B_1 = mq\eta x/4\pi R^3r^3 \quad (8)$$

If  $F$  be the force per unit volume at  $P$ , we have, by (8),

$$F = \frac{mq\eta}{4\pi R^3} \frac{d}{dt} \left( \frac{x}{r^3} \right) = \frac{mq\eta}{4\pi R^3} \frac{d}{dx} \left( \frac{x}{r^3} \right) \quad (9)$$

since  $dx/dt = u$ .

Now

$$\frac{d}{dx} \left( \frac{x}{r^3} \right) = \frac{-2x^2 + xR \cos \theta + R^2}{r^5} = -\frac{1}{R} \frac{d}{dR} \left( \frac{R^2}{r^3} \right) \quad (10)$$

and hence

$$F = -\frac{mq\eta}{4\pi R^4} \frac{d}{dR} \left( \frac{R^2}{r^3} \right) = -\frac{mq\eta \sin \theta}{4\pi R^3} \frac{d}{dR} \left( \frac{R^2}{r^3} \right) \quad (11)$$

This force acts in circles about  $OX$ , and thus, if  $L$  be the moment about  $OX$  of the forces on the medium between the two spheres,

$$\begin{aligned} L &= \int_a^b \int_0^\pi F \cdot 2\pi R^3 \sin^2 \theta dR d\theta \\ &= -\frac{mq\eta}{2} \int_a^b \int_0^\pi \frac{d}{dR} \left( \frac{R^2}{r^3} \right) \sin^3 \theta dR d\theta \\ &= -\frac{mq\eta}{2} \int_0^\pi \left( \frac{b^2}{r_b^3} - \frac{a^2}{r_a^3} \right) \sin^3 \theta d\theta \end{aligned} \quad (12)$$

where

$$r_a^2 = x^2 - 2xa \cos \theta + a^2$$

$$r_b^2 = x^2 - 2xb \cos \theta + b^2$$

Then, by (5),

$$L = -\frac{2mq\eta(b^2 - a^2)}{3x^3} \quad (13)$$

Hence  $L = -J$ , and the resultant moment of the forces on the charges and of the forces on the medium vanishes.

Although the moving pole creates no resultant torque, it has the remarkable effect of urging the combined pair of spheres to turn in one direction and the medium between them to turn in the opposite direction.

An isolated pole does not exist. If we have a magnet lying along  $OX$ , with a pole  $+m$  at distance  $x$  from  $O$  and a pole  $-m$  at distance  $x + l$ , the resultant moment on the pair of spheres is

$$\frac{2}{3}mq\eta(b^2 - a^2) \left[ \frac{1}{x^3} - \frac{1}{(x + l)^3} \right]$$

## NOTES AND NEWS

## New Books

**The Properties of Metallic Materials at Low Temperatures.** By P. LITHERLAND TEED. (London: Chapman and Hall, Ltd.) Pp. viii + 222. Price 21s. net.

This book is the first of a series of monographs on metallic materials to be published under the authority of the Royal Aeronautical Society. These volumes are evidently intended to survey, primarily for the benefit of aircraft designers, existing information on the mechanical properties of the materials which metallurgists offer them.

The author has cast his net wide and gathered a surprisingly large volume of data. In 45 tables he sets out, for a wide range of aluminium alloys, plain and alloy steels, magnesium, copper, nickel, zinc, tin and lead-base alloys values, at temperatures between  $+20^{\circ}\text{C}$  and  $-270^{\circ}\text{C}$ , for the yield stress, ultimate tensile stress, elongation and reduction of area, Young's modulus, fatigue endurance limit, notched-bar impact strength and electrical resistivity. This conveniently tabulated information cannot fail to be directly useful, after the manner of the results obtained in conventional mechanical tests, to those concerned with designing structures for low-temperature service. Moreover, they will find it easy, if they wish, to discover the source of any item of tabulated information.

The special importance which designers of high-altitude machinery must attach to the effects of low temperatures is evident, and it is appropriate that the first monograph of the series should review this aspect of the strength properties of constructional alloys. Opportunities for discussing the engineering significance of the results which are quoted are however, limited, since they show that, in general, all the technical strength values reviewed are somewhat enhanced by reduction of temperature. Important exceptions are provided by ferritic plain-carbon steels and by zinc base alloys, the notched-bar impact resistance of which decreases sharply with fall in temperature.

The author therefore confines himself mainly to explanatory remarks on the tables, referring, where he has found it possible, to work bearing on the reasons for the observed effects of temperature change.

G. L. J. BAILEY

**The Physics of High Pressure.** By P. W. BRIDGMAN. (London: G. Bell and Sons, Ltd.) Pp. vii + 445. Price 35s. net.

This comprehensive treatise is a worthy addition to that excellent series "International Text Books of Exact Science" edited by Professor E. N. da C. Andrade. Professor Bridgman has achieved an international reputation for his researches on the physics of high pressure and this book gives an authoritative account of the subject, particularly of the author's own work.

This volume, which has an excellent bibliography, contains a reprint of the original book published in 1931, with one main modification, together with a supplement covering the work carried out since then. The first chapter gives an historical account of the development of this branch of physics. Although work is still confined to a relatively few laboratories there has been accelerated activity in the high pressure field since 1931.

The second and the third chapters describing high pressure technique are of considerable interest. The discovery of the

principle of the unsupported area of packing by Bridgman opened up the whole field of high pressure physics. He first increased the pressure obtainable to  $12\,000\text{ kg/cm}^2$ , and since 1931 to  $100\,000\text{ kg/cm}^2$ . It should be pointed out, however, that the pressures are only truly hydrostatic up to  $30\,000\text{ kg/cm}^2$ .

Chapter IV is concerned with the effect of pressure on the plastic properties and fracture of solids. It is interesting to note that the shearing strength of metals is not independent of the normal stress on the slip planes under high pressure. The remaining chapters are devoted to the effect of pressure on such physical properties as the *P.V.T.* relations in fluids, compressibility, melting phenomena, resistivity and thermal conductivity. Some idea of the scope and wide appeal of the book may be obtained from this selection of chapter headings.

Apart from some minor printing errors there is little to criticize in the book. However, the frequency with which units are omitted is a little disconcerting, e.g. in quoting pressure coefficients, and absolute viscosities, and in many cases pressures are given in kg. Larger illustrations and more detail in the description of techniques would have been welcomed, but perhaps it was impossible to incorporate such information.

The book must be regarded as a standard work on the experimental side of high pressure physics. It will be welcomed both by those interested in the physical properties discussed and by those concerned with the application of high pressures.

H. LL. D. PUGH

**The Mathematical Theory of Communication.** By C. E. SHANNON and W. WEAVER. (Urbana: University of Illinois Press.) Pp. 117. Price \$2.50.

The general theory of communication means a consideration of the various sources of information, the capacity of transmitters in expressing messages by signals, the capacity of a channel and receiver to pass the signals to a distant point and to reconstruct the original messages, and finally the destined recipient which has to react to the intention of the source of information; but this last element does not really enter into the present book. The mathematical basis resembles that of other kinds of operational research, although operational research is so inherent in applied electricity that it is rarely so called. In the present instance the breadth of the basis may be illustrated by the less serious by-product of the adaptability of languages to cross-word puzzles. The book is in two parts, and the second surveys the broad field just mentioned. The authors suggest that some readers may prefer to start with the second part, and this seems so true as to cause wonder at the order of printing. Unfortunately, an attempt has been made in the second part to help the less expert reader by explanations and analogies which will confuse and, if a physicist, annoy him. The first part contains a mathematical analysis, leading to twenty-three theorems, and some detailed expansions and proofs in seven appendices. Particular attention is given to the influence of noise, in a general sense, in degrading the range and reliability of a system. The treatment begins with sources of information which give rise to discrete sequences of symbols which can be encoded as such. One example is a language message



here the letters have individual probabilities, two or more letter sequences have certain probabilities or, taking a further step, words have a statistical structure in the language. Another example is the numerical information fed into a computing machine or repeater. Noise introduces uncertainty or equivocation. These sections are treated with some mathematical rigour. Less rigour is practicable or necessary in dealing with the transmission of continuous functions and the effect of noise thereon. These functions are of increasing importance in telecommunications, including television and radar, and some forms of computation (e.g. analysers and predictors). Apart from setting out the general theory, emphasis is laid on the matching of the statistical properties of the component parts of a system and the theoretically possible rates, with and without noise, at which information can be sent correctly. This book cannot be ignored by anyone with direct professional concern with these applications and many applied physicists without this concern should, like the reviewer, find the book absorbing. S. WHITEHEAD

**Practical Applications of Spectrum Analysis.** By H. DINGLE, D.Sc. (London: Chapman and Hall, Ltd.) Pp. ix + 245 + xix plates. Price 40s. net.

Practical spectrographers will be disappointed with this book, particularly after reading the statement in the Preface: "The purpose of this book is to provide a trustworthy guide, as complete and self-contained as is possible in a single volume, for the practical spectroscopist engaged in analytical work connected with any field of study whatever."

The chapter on "Sources of Luminosity" is sadly out of date. No mention is made of the Lundegårdh flame apparatus, modern arc sources (d.c. and a.c.), uncontrolled and controlled spark discharges or multi-source units, the successful applications of which are common knowledge to practical spectroscopists.

Professor Dingle considers that problems of spectrographic analysis should be attacked by a theoretical approach "rather than by narrowly empirical methods which . . . have no trustworthy wider application." Whilst we all agree on the desirability of a method of general applicability, based on theoretical grounds, in the present state of knowledge of the subject the view of Professor W. E. S. Turner (*Physics in Industry*, Lecture No. 12, 1927) surely represents a more enlightened attitude: "When scientific workers are apt to carry sometimes to excess their criticism of empiricism in industry and be dubious of the possibility of any progressive creative factor other than that which they carry into field and factory, it serves perhaps as a little corrective to be reminded that epoch-making developments may still arise through the imagination, the industry and determination of, scientifically, unlettered men."

Only  $7\frac{1}{2}$  pages (Chapter 8) are devoted to quantitative analysis and the only analytical method discussed is that of E. W. Foster. Quoting again from the Preface: "The small amount of space given to quantitative spectrum analysis properly reflects, I believe, the present state of achievement in this field, but not the promise for the future." Much work in devising accurate quantitative methods of analysis, of high scientific value, has been initiated and carried out in industrial spectrographic laboratories, both in this country and abroad, and it is discouraging, to say the least, to have such work so lightly dismissed; in fact, ignored.

The remaining chapters, dealing with general principles,

prismatic and diffraction grating spectrographs, the determination of wavelengths, and qualitative analysis, are concise and authoritative. A very useful feature of the chapter on qualitative spectrum analysis is the discussion of the most persistent lines of the elements and their multiplet relationships. Possible coincidences with lines of other elements are noted.

The Appendix (containing eight Tables) occupies 115 pages, and the book concludes with 19 plates of reproductions of spectra. Table V combines the lists of persistent lines from the M.I.T. Wavelength Tables and the Hilger tables, and Tables VII and VIII record the wavelengths and identifications of the lines appearing in the spectra of the "raies ultimes" mixture (Plates I-V, VII-VIII, with juxtaposed iron spectra). This mixture was made at the Imperial College laboratories and is more comprehensive than the "R.U. Powder" made in the Research Laboratories of the General Electric Co., Ltd.

In fairness to the author it should be pointed out that he states that "the book is essentially a record of personal experience rather than a general conspectus of all the methods that have been used." It would have been preferable, therefore, to have qualified the title in some way to give a clearer indication of the contents. D. M. SMITH

**The Practice of Electron Microscopy.** Edited by D. G. DRUMMOND, Ph.D., for the Electron Microscopy Group of The Institute of Physics. (London: The Royal Microscopical Society.) Paper covers, pp. 141. 22 plates. Price 21s. including postage.

A group of twelve electron microscopists, members of the Electron Microscopy Group of The Institute of Physics, have combined under the editorship of Dr. D. G. Drummond to produce this comprehensive treatise on the detailed techniques used in the preparation of specimens for examination in the electron microscope.

Care has been taken to give full working details of the techniques even to the extent of enumerating the pieces of apparatus required, carefully identifying proprietary materials and naming the suppliers. The book contains many previously unpublished communications and gives full references to originators of particular techniques.

This work will be of great value to all new and potential electron microscopists who will now be able to draw more readily on the experience of those who have learned "the hard way." The treatise will be no less valuable to the experienced electron microscopist who, in this age of specialization, cannot hope to have intimate knowledge of the ultimate practical details of all the techniques.

The book is extremely well produced and contains many excellent reproductions of electron micrographs. It is perhaps unfortunate that the legends for the electron micrographs do not appear on the micrographs themselves but are collected together at the end of the treatise. There are three additional chapters on instrumental questions and four appendices.

The end of the Foreword may be quoted: "It is believed that Dr. Drummond will have the satisfaction of seeing his work become an invaluable reference book for electron microscopists, experienced and inexperienced alike, and an object of comparative study for microscopists generally."

F. W. CUCKOW

*Members of The Institute of Physics are entitled to one copy each of the above book for their personal use at 10s. 6d., including postage, if ordered through the Institute.*

## Notes and Comments

### The Institute of Physics

At the Annual General Meeting of The Institute of Physics, held on 20 July, 1950, Prof. W. E. Curtis (Professor of Physics, King's College, University of Durham, Newcastle-upon-Tyne) was elected President.

Prof. E. G. Cox was elected a Vice-President, and Dr. F. P. Bowden and Mr. G. R. Noakes were elected Ordinary Members of the Board. The Honorary Treasurer, Mr. E. R. Davies, and the Honorary Secretary, Dr. B. P. Dudding, were re-elected. Prof. H. W. Thompson joins the Board as the new representative of the Faraday Society.

The Report for 1949 which was adopted at the meeting records continued progress in activities and in numbers. During the year the Education Group and the North-Eastern Branch were inaugurated, making the seventh specialist subject Group and the eighth Branch. These fifteen Groups and Branches held many meetings and visited various laboratories; the five Divisions of the Australian Branch held 39 meetings during the year.

Nine colleges in England and Wales have had their courses approved for National Certificates in Applied Physics and arrangements were made with the Scottish Education Department for the establishment of similar courses in Scotland.

The Institute collaborated with the Universities of Bristol and Cambridge and University College, London, in Summer Schools on special branches of physics.

The Report records the decision to establish the *British Journal of Applied Physics* and a monthly *Bulletin*. A further volume was added to the "Physics in Industry" Series. A second edition of the *Handbook of Industrial Radiology* was published during the year and also a selection from the *Laboratory and Workshop Notes* which had appeared in the *Journal of Scientific Instruments*.

The Institute's Officers have dealt with many inquiries about salaries, conditions of service, consulting fees, professional training and similar matters. During the year the Board had before it items concerning certain Bills, then before Parliament, and inquiries from Government Departments concerning physicists and training in physics.

The Report shows that the Institute continues to be represented on many committees and to collaborate closely with its sister Institutes in various activities.

### Certificates for Laboratory Technicians

The Board of The Institute of Physics announces that the Institute's Certificate in Laboratory Arts will not be awarded after 1951, in view of the establishment of the Intermediate Certificate for Laboratory Technicians by the City and Guilds of London Institute, which it welcomes.

For some time it has been felt desirable that there should be established a Certificate which would be of value to technicians in a wider range of laboratories. The Institute of Physics was represented on the Exploratory Committee of the City and Guilds of London Institute whose work has resulted in the publication of regulations and syllabuses which are coming into operation in the session 1950-1951.

The City and Guilds of London Institute has agreed that holders of The Institute of Physics' Certificate shall be eligible to enter, should they so wish, the course for the Advanced Laboratory Technicians' Certificate which is to be established

shortly on the same terms as those who have passed examinations for the new Intermediate Certificate and this is regarded as a fitting recognition of the standard which The Institute of Physics' Certificate holders had reached and of the pioneer work which that Certificate represented. Students who have already begun courses for The Institute of Physics' Certificate will be able to complete these by 1951 after which The Institute of Physics will no longer examine in Laboratory Arts.

The Board of the Institute has formally recorded its appreciation of the important work which has been done by those who have established and taught the courses for its Certificate and hopes that their work will be continued under the new scheme.

## Journal of Scientific Instruments

### Contents of the October issue

- A Mechanical X-ray Structure Factor Calculating Machine. By V. Vand.
- The Properties of Proportional Ion Chambers with Glass Envelopes and External Graphite Electrodes. By A. H. Cockroft and J. M. Valentine.
- Field Measurements on Model Betatron and Synchrotron. By E. A. Finlay, J. F. Fowler and J. F. Smee.
- A Photoelectric Polarimeter. By D. H. Rank, J. H. Light and P. R. Yoder.
- The Conifuge—a size separating sampling device for airborne particles. By K. F. Sawyer and W. H. Walton.
- A Simple Machine capable of Fourier Synthesis Calculations. By J. H. Robertson.
- An Instrument for directly recording the Metacentric Height of a Ship. By A. G. Boggis and E. C. B. Corlett.
- A Small High-temperature High-vacuum Furnace. By K. H. Alberman.
- A Nickel Chromium-Iron Alloy for Sealing to Glass. By J. E. Stanworth.

### NOTES AND NEWS

#### *New Instruments, Materials and Tools.*

Angle Calculator—Mercury-in-Glass Thermoregulator—Recording Dew-point Hygrometer—Insulation Tester—Reed Vibrometer Amplitude Scale—Insulation Test Set—Automatic Shaft-setting Mechanism—Reflecting Microscope Objectives—Germanium Crystal Rectifiers

#### *Notes and Comments.*

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## SPECIAL REPORTS

## Conference on Vacuum Physics—Birmingham, 1950

The Midland Branch of The Institute of Physics held a conference on "Vacuum Physics" in the University of Birmingham on 27 and 28 June, 1950; and a small exhibition was held at the same time. The Conference served the two purposes of a review, for industrialists, of the present position in the various fields of vacuum technique and engineering, and an opportunity for presentation, and discussion, of original work, by those actively engaged in research in this field. The papers presented and the discussion on them are summarized in this report. A brief account of the exhibition is also included.

The conference was opened by PROF. OLIPHANT, who stressed the importance of such problems as uniformity in vacuum "plumbing," provision of baffles which do not reduce pumping speed substantially, the need for gauges which will reliably record the true pressure, and cheaper pump oils which do not decompose into a "tar," which in time makes the research worker's apparatus inoperable. Two anecdotes demonstrated admirably how the provision, or not, of a good vacuum, can mean success or failure to a nuclear physicist, whose experiment is otherwise planned on the soundest physical bases. Means of obtaining and measuring a vacuum are rather different to-day from those once employed, such as calculating pumping speed and pressure in terms of the geometry and number of cycles of a Töpler pump.

The reviews of modern vacuum pumps and gauges given by MR. D. GODDARD, of British American Research Ltd., and MR. STECKELMACHER, of W. Edwards and Co. (London) Ltd., showed that a wide range of mechanical and diffusion pumps and of gauges is now available in this country.

Recent additions are a robust magnetically damped Knudsen gauge, and a sensitive cold-cathode ionization gauge useful down to a pressure of  $10^{-6}$  mm of mercury, both now available commercially. The higher pressure gauges are useful for protective and automatic control of industrial plants. Pump manufacturers now provide efficient baffles, traps, and high vacuum valves, with their equipment. The application of this equipment to the design of industrial vacuum systems was discussed by MR. J. MILLEN, of Metropolitan-Vickers Electrical Co. Ltd. Vacuum systems are no longer of a laboratory experimental nature. The problems to-day are engineering ones, and careful selection of components and materials of construction is necessary, with emphasis in industry placed upon reliability of operation. Any design depends on the specific application, and generally a compromise has to be reached between the several factors involved. A group of smaller pumps is to be preferred to one very large one, and emphasis is placed on the need for testing sub-assemblies. The provisions of sufficient vacuum valves, a by-pass line, and automatic protection, control, and signalling are very important.

Intimately related with testing is, of course, the problem of leak detection. A paper on the general principles of leak-detection, with special reference to the differential Pirani and ionization gauge leak-detectors, was read by MR. J. LECK and MR. J. BLEARS, both of Metropolitan Vickers Electrical Co. Ltd. Both detectors employ two gauges balanced in a bridge circuit, so that background fluctuations are small. In the Pirani detector a calcium hydroxide trap is placed in the line to one gauge, so that using carbon-dioxide as probe gas, an out-of-balance current is recorded upon finding a leak. The sensitivity limit is less than  $10^{-3}$  litre-microns per second (or lusec). For ionization gauges with a liquid-air trap and butane as probe gas, the minimum leak detectable is of order  $10^{-6}$  lusec.

This figure compares very well with the helium mass spectrometer leak detector, described by DR. C. MILNER, of the British Thomson-Houston Co. Ltd. With a cold-cathode ion source, the limit with the latter instrument is rather less than  $10^{-5}$  lusec, and with a hot-filament type of source, a leak of  $10^{-7}$  lusec is detectable. The choice of a particular method of leak detection depends on the problem in hand. For industrial applications such as sealed-off valves and metal-rectifiers, which have to operate for many years, there is room for still better leak detectors than are at present available, particularly in respect of probing methods for pin-pointing leaks more exactly. For large dynamic systems which cannot be outgassed, a leak-rate of the order of one lusec is satisfactory, and in this case a single non-stabilized ion gauge is satisfactory.

MR. L. RIDDIFORD, of the University of Birmingham, said that the detection of leaks of 0.01 lusec is possible using liquid trichlorethylene, for which the amplification factor over air is about 25. Liquid probing with a painter's brush permits accurate pin-pointing of leaks.

Mr. Riddiford, in his paper on the vacuum systems of particle accelerators, stressed that modern vacuum engineering has arisen primarily on account of the demands of the physics research laboratory. The giant synchro-cyclotrons and proton synchrotrons to-day in operation, or under construction, require pressures as low as about  $10^{-5}$  mm of mercury or less for successful operation, since gas scattering causes loss of beam to

the walls of the vacuum box. In the 1 300 MeV synchrotron in the University of Birmingham, a pressure of  $10^{-6}$  mm of mercury is required for certainty in operation.

The vacuum chamber is a hollow ring 30 ft in diameter, with rectangular section 40 cm  $\times$  10 cm, constructed from 30 ceramic sections each about 1 m long. It is evacuated by five 16-in diffusion pumps, at the head of each of which is a stainless-steel manifold. The nature of the pump ultimate molecules is important since, for example, one oil molecule "scatters" as effectively as 13 air molecules. Evidence indicates that they are, in fact, oil decomposition products of average molecular weight about 160.

More detailed information on this point was presented by MR. BLEARS, in a paper on studies in high vacua with the mass spectrometer. The mass-distribution curve is almost continuous up to a mass number of 300. Further important results obtained with the spectrometer, are that water vapour molecules are predominant during initial evacuation of a vacuum vessel, and that mercury pumps behave in the same manner as oil pumps, owing to the presence of backing pump oil molecules in any system.

One session was devoted to an interesting series of short talks on vacuum plants to-day operating in industry, and the importance of their application. MR. L. HOLLAND, of W. Edwards and Co. (London) Ltd., discussed recent advances in vacuum coating plant and technique, with particular emphasis on the importance of rapid pumping cycles, of order 15 min, which have only been made possible by recent pump developments. The problems peculiar to the melting and sintering of metals in vacuo were described by MR. E. MALCOLM, of British American Research Ltd. Various systems for degassing, melting, and casting 50 to 100 lb of steel, were discussed. The importance of centrifugal freeze drying to medical research and practice, was emphasized by MR. L. BECKETT, of W. Edwards and Co. (London) Ltd. Biological products are dried in the frozen state, and subsequently concentrated or preserved or both. Provision of a vacuum is essential in the drying of heat-sensitive materials, and when large quantities are to be treated, the problem is one of large-scale vacuum dehydration. Recent plants constructed for this pur-

pose were described by MR. D. GODDARD, of British American Research Ltd.

One of the most interesting exhibits in the exhibition was a large pumping manifold, and sections of the vacuum chamber, of the 1 300 MeV proton synchrotron, together with several other items of equipment employed in the vacuum system of that particle accelerator, exhibited by the University of Birmingham.

Three firms, namely British-American Research Ltd., W. Edwards and Co. (London) Ltd., and Metropolitan-Vickers Electrical Co. Ltd., exhibited a wide range of vacuum equipment. Mechanical pumps of all sizes up to the Kinney range, and diffusion pumps of diameters ranging from 1 in to 16 in, were to be seen. Related to these are baffles and plate valves of corresponding sizes, in one case the two features being included in the one plate and housing. A powerful 6-in booster pump, and a visual demonstration of the jets of a 6-in metal pump in operation, were added features of the pumping display. A complete range of high- and low-pressure gauges, with their associated circuits and controls and protective devices derived from the gauges, was shown. Of special interest were alphasatron, Knudsen, and cold-cathode ionization gauges. Leak-detectors consisted of balanced thermocouple and ionization gauges, and the hot filament  $90^\circ$  helium mass-spectrometer.

The British Thomson-Houston Co. Ltd. displayed their  $180^\circ$  mass spectrometer leak detector, which is mounted in an accessible-compact cabinet, on wheels, a feature attractive to industry. Commercial auxiliary equipment shown included valves, both manually and automatically operated electrically or pneumatically, water switches and flow-meters, cold-traps, and various connexions, fittings and gaskets essential to sound vacuum "plumbing."

Examples of metal-coated materials such as plastics, reflectors, prisms and lenses, and paper, were exhibited, together with the coating plants. Completely automatic and compact cathode-ray tube pumping plants were displayed and a large twin-tank high vacuum coating unit was shown in operation. A large automatically operated discharge tube demonstrated the reliability of present control systems.

L. RIDDIFORD.

It is intended that the papers and the discussion on them shall be published in a special issue of the *Journal of Scientific Instruments* next year. This is in place of publication in the Institute's "Physics in Industry" series as previously announced.



## Summarized Proceedings of a Conference on the Physics of Lubrication—Manchester 1950

A joint Conference of the Manchester and District Branch of The Institute of Physics and The British Rheologists' Club was held at The University of Manchester from 29 June to 1 July, 1950. The first session was devoted to "Hydrodynamic lubrication and the rheology of lubricants," the second and third sessions to "Boundary and extreme pressure lubrication."

The papers and the discussions on them are summarized in this report.

In opening the Conference, PROFESSOR F. A. VICK, the Chairman of the Branch, said that the University of Manchester was an appropriate place for the Conference, since it was here that Osborne Reynolds did his fundamental work and was active for a long time. The purpose of the Conference was to review the physical aspects of lubrication and to discuss recent advances in this field.

The attendance of some 200 people and the liveliness of the discussions showed the widespread interest in the subject among scientists and engineers, and in view of this it is tempting to speculate on why so little fundamental work on the subject is being done at present. It offers a fascinating field to any critical and painstaking physicist who cares to study it.

### HYDRODYNAMIC LUBRICATION AND THE RHEOLOGY OF LUBRICANTS

PROFESSOR D. G. CHRISTOPHERSON (University of Leeds) began his "Survey of Hydrodynamic Lubrication" with Osborne Reynolds' interpretation of Beauchamp Tower's experiments enunciating for the first time in 1886 the principle that the load-bearing capacity of a fluid-film depended upon the pressures developed when the lubricant was drawn through a constriction or convergent wedge-shaped space under the action of viscous forces. The assumptions on which Reynolds' paper was based may be divided into two classes, those which were fundamental and without which any progress could have been made on the basis of general hydrodynamic theory only with extreme difficulty, and those which were useful in that they simplified the mathematical work, but which were not nearly so essential. There is first an assumption which does not fall clearly into either of these categories, and this is that fluid velocities perpendicular to the solid surfaces bounding the lubricating film are negligible compared with those in the plane of relative motion, and a related assumption, that the velocity gradient through the film is very large compared with other velocity gradients. The essential assumptions are (1) that the inertia forces, owing to the density of the lubricant, are negligible; (2) that the behaviour of the lubricating fluid can be completely defined in terms of its bulk properties, viscosity, density, and that these quantities are single-valued functions of temperature and pressure; (3) that the geometry of the bounding surfaces is completely known beforehand. It is also an almost essential assumption that the temperature should not vary through the thickness of the

film. Assumptions (2) and (3) define the limits within which hydrodynamic theory is applicable in the course of the transition to the thin-film region. If, under practical conditions, there is a layer of lubricant on each surface in which quite different considerations apply, and if, as seems probable under conditions of heavy load, microscopic changes are continually taking place in the geometry of the solid surfaces, it is only when one considers films which are thick compared with the surface layers, and regions which are large in comparison to the deforming area, that the hydrodynamic theory can be applied. Among the inessential "simplifying" assumptions is the question of side leakage. Professor Christopherson referred to the pioneer work of Michell,<sup>(1)</sup> the experimental studies of Kingsbury,<sup>(2)</sup> the extensive computations of Muskat and Morgan,<sup>(3)</sup> and the numerical methods employed by Cameron and Wood<sup>(4)</sup> which have between them provided valuable, if incomplete, information relating the load-carrying capacity and frictional force of a bearing to its length/width ratio.

The temperature of the fluid film is important as it affects the viscosity and gives rise to volume expansion (the so-called "thermal-wedge" effect) which is probably partly responsible for the satisfactory functioning of the high-speed parallel-faced thrust bearing, as observed by Fogg.<sup>(5)</sup> Many attempts have been made to determine theoretically the temperature distribution over the bearing surface (Cope).<sup>(6)</sup> Little progress has been made as yet with regard to the question whether the geometrical changes caused by temperature gradients in the metal parts of the bearing do or do not have any important effect on performance.—There is still much work to be done to determine the variation of viscosity with pressure for lubricants of practical importance. Professor Christopherson considered that good "oiliness"—which in a symposium of physicists he would define as that quantity which determines the lubricating behaviour of an oil when the load is increased or the speed diminished until hydrodynamic lubrication has broken down—and a high pressure-coefficient of viscosity go together in practice. He made, however, the reservation that other properties such as boundary lubrication properties also enter, and these seem to have little to do with the pressure-coefficient of viscosity. He did not wish to anticipate anything connected with the viscosity of lubricating fluids at high rates of shear as there was to be a paper on this subject during the Conference.

Hydrodynamic lubrication will break down at those places and times when two asperities of the solid surfaces happen to be exactly opposed. Thus, if the coefficient of boundary friction is known, it is not difficult to deduce the so-called contact-load ratio from observations of the actual force of friction in the bearing combined with a knowledge of the theoretical force of hydrodynamic friction. There are, however, complicating aspects. Firstly, even before boundary lubricating conditions can be established near the peak of an asperity the pressure may exceed the yield stress of most bearing metals when the film thickness is still substantially more than what one normally considers to be the thickness of a boundary lubricating film with the ensuing flattening of the asperity. Secondly, the high pressure is produced only on the upstream face of the asperity with a pressure reduction on the downstream face. There is no doubt that a great deal of smoothing does go on in the running-in process.

Apart from the effect of load in changing microstructure there may be a much larger effect in deforming the geometry of the bearing as a whole. For example, a shaft running in a complete journal bearing will tend to dig a "pit" for itself with the result of increasing the load-carrying capacity of the bearing by increasing the "conformity radius" so that a bearing which is always loaded in the same direction running at first under boundary conditions may "run in" to a state in which the load is carried hydrodynamically.

Cavitation phenomena in the negative pressure region have been studied by using transparent bearing materials. What happens in the cavitating region is still indefinite. In the normal course of events a particular bubble will travel round the bearing until it comes to the place at which the pressure begins to increase. It will then recondense. But suppose that the load on the bearing is pulsating and that therefore a bubble finds itself suddenly in a region of high pressure. Such conditions would be ideal for the occurrence of cavitation erosion. Apart from the "bulk" cavitation which may occur over an appreciable part of the bearing arc, there is the possibility of local minute cavitations taking place on the downstream side of the microscopic surface irregularities. The formation of such small cavities in a region of general positive pressure would be resisted by surface tension and it may be that they do not occur, but are replaced by some form of eddy flow.

Professor Christopherson concluded by trying to guess which of the developments now taking place will appear to an observer twenty years hence, to have been of the greatest importance. On the engineering side of fluid lubrication only, two related events stand out. It has become more widely realized that careful attention to the question of ensuring that hydrodynamic lubrication obtains wherever possible will give very great returns by way of reduction of wear and longer life, and the necessary knowledge both in design and in questions as the selection and treatment of lubri-

cants is becoming much more widespread. Perhaps arising out of this increased appreciation by industry there is an ever-increasing tendency to demand that more and more types of rolling and translational contact should be brought within the range of fluid lubrication.

In the discussion, Dr. G. W. NEDERBRAGT (Koninklykel Shell Laboratorium, Amsterdam) asked about the influence of the molecular constitution of the lubricant on its thermal conductivity. PROFESSOR CHRISTOPHERSON considered that the heat transfer by convection was more important in a bearing than that by conductivity. MR. H. BLOK (Koninklij/Shell Laboratorium, Delft) mentioned the "circular hill versus ridge" aspect of the bearing surface in connexion with the suggestion of a partial flattening of the asperities of the surfaces even under conditions of hydrodynamic lubrication. He referred to Ladanyi's work as indicating that the pressure transmitted through the lubricant would tend to flatten sharp peaks to form a series of plateaux. On the other hand, if the original surface irregularities were of the type of circular hills instead of tall peaks very slight flattening would appreciably reduce the pressure required to maintain plastic deformation. COMMANDER J. R. D. WALKER (Research Department, Vickers-Armstrongs Ltd.) inquired about the correlation between film thickness and solidification.

DR. E. W. J. MARDLES (Royal Aircraft Establishment, Farnborough) gave a paper on "The rheology of lubricant films." He presented in condensed form many results of studies of the rate of radial flow over horizontal flat polished surfaces and between optical flats that indicate the adhesiveness of thin layers of liquids. Under pressures much smaller than are normally used in friction experiments under boundary lubrication conditions, the influence of the boundary lubricant layer in supporting the floating solid body appears to be extended into the bulk of the liquid. Dr. Mardles had investigated with several liquids at pressures less than 1 g-wt/cm<sup>2</sup> deviations from Amontons' law such as were reported by Sir William Hardy,<sup>(7)</sup> and he ascribed the deviations to surface forces affecting the liquid between "points of contact." In pulling apart two flat surfaces there appeared to be a latent period, then some movement and breaking away.

A joint paper by DR. A. F. H. WARD, DR. S. M. NEALE, and MR. N. F. BILTON (Faculty of Technology, University of Manchester) on "The viscosity of lubricating oils at high rates of shear" was presented by Dr. Ward. He described in detail a high-speed rotational viscometer with slightly conical stator and rotor and with a very thin oil film. The rotor and stator were allowed to centralize themselves, and the torque was measured with two spring balances. Thermocouples were used to indicate the temperature of the stator at various points, and the temperature readings were accurate to 0.5° C. Special attention was given to the estimation of the temperature of the oil film so that the fall in viscosity caused by the rise in temperature might



was allowed for. It was then found that at high rates of shear the coefficient of viscosity showed a decrease which was a function of both the rate of shear and the temperature. The accuracy of the viscosity measurements was  $\pm 4\%$  mainly owing to a 2% error in determining the magnitude of the clearance. The calculated viscosity was a mean value because the temperature of the oil film varied by as much as 20°C across the film. The assumption was made that little heat was lost from the rotor so that its temperature equalled that at the inner surface of the oil film. The temporary viscosity reduction was assessed by heating the oil in a low rate of shear viscometer. As in this subsidiary experiment a temperature had to be reached that was above the temperature of the rotor in order to obtain similar apparent viscosities in both experiments, the conclusion was drawn that, in the rotational viscometer, a viscosity reduction exceeding the temperature effect had been achieved as a result of the rate of shear. The results obtained with the apparatus could thus be interpreted in terms of a balance between the flow orientation produced by the velocity gradient, as predicted by Grunberg and Nissan,<sup>(8)</sup> and the disorientation caused by Brownian movement as discussed by Robinson.<sup>(9)</sup> A much more marked effect was observed when long-chain molecules (viscosity-index improvers) were added to the oil.

In reply to a question by DR. MARDLES, DR. WARD said that no permanent viscosity loss, as distinct from a temporary viscosity reduction owing to flow orientation, had been found with the apparatus when polyisobutylene had been dissolved in the oil. One speaker mentioned the possibility of a hump in the curve of the temperature gradient across the oil film and the effect on the temperature distribution of the rotation of the inner cylinder. DR. NEALE wound up the discussion on this paper expressing the hope that in view of the difficulties encountered with the application of rotation viscometers to high rate of shear measurements further work would be undertaken on similar lines.

The last paper in this Session was given by MR. E. MOSS (British Cotton Industry Research Association, Manchester), who spoke on "The lubrication of textiles." He said that the yarn friction was important in the spinning, weaving, knitting and stitching operations. The force of friction of a thread passing a cylindrical guide was  $\exp(\mu\theta)$  where  $\mu$  is the coefficient of friction and  $\theta$  is the angle subtended by the area of contact. At a speed of 60 yd/min the coefficient of friction of a natural yarn fibre practical operating conditions (i.e. tension on the fibre) was 0.25. Most fibres contain a natural lubricant. For instance, cotton wax was the natural lubricant of cotton fibres. But when this greasy layer was removed the coefficient of friction might increase to a value of 0.7. This is the stage at which under practical conditions serious difficulties arise. The view was expressed that the force of friction rises when the layer of wax molecules becomes 2 to 3 molecules thick. Dyeing can increase the fric-

tional force, and the effect of twist on friction was important. A low and uniform force of friction was required for knitting. The force of friction was diminished to different extents by alcohols, organic acids, soaps and paraffin wax.

In the discussion on this paper inquiries were made about the value of the coefficient of static friction and of the effect of speed on the frictional force, on the contribution of electrostatic charges to textile friction, on the effects of humidity and of the quality of the surfaces of cylindrical guides (i.e. the effect of glazing).

Mr. Moss exhibited two pieces of laboratory apparatus which are used for the measurement of textile friction.

#### BOUNDARY LAYER AND EXTREME-PRESSURE LUBRICATION

DR. D. CLAYTON'S (Imperial Chemical Industries Ltd.) survey on "Boundary layer and extreme-pressure lubrication" introduced the second part of the Conference. Dr. Clayton sketched the general background of his subject chiefly for those not engaged in investigations of boundary lubrication so that they could see this subject in perspective and also see where individual contributions fitted into the general picture. He therefore did not strive to include the latest results that have been published, trusting that the discussion would reveal the trend of events. Complete boundary lubrication and complete fluid-film lubrication were, he said, extreme cases and there were many mechanisms in which there was a mixture of the two regimes, at least at some period. The designer's aim should be to ensure that a fluid film was obtained because the force of friction was then low and ideally there was no wear. However, wear could occur even with nominally thick-film lubricated surfaces if particles (air-borne dust, for example) were present that are larger in dimensions than the thickness of the film. Boundary lubrication was essentially a function of both the solid materials and the lubricant. At one time it was thought that a lubricant would have a constant friction-reducing effect. This was based on the idea of an adsorbed film of lubricant which would satisfy the surface forces and separate the surfaces by an amount proportional to the size and in particular to the length of the molecule. Friction was explained in the early days by the interlocking of surface asperities (Coulomb). More recently an adhesion hypothesis by which friction is explained in terms of molecular adhesion between the solid surfaces has been proposed. Dr. Clayton mentioned as an example of the latter that if perfectly clean metal surfaces were brought together they would stick, and he referred to Bowden and Hughes<sup>(10)</sup> and Bowden and Young's<sup>(11)</sup> reports of coefficients of friction of anything from 5 to 100. Mention was made of the late Dr. Tomlinson's<sup>(12)</sup> experiments with a three-legged friction element with lead feet which would stick so firmly to a "cleaned" glass plate that the plate could be inverted without the other friction element falling away. The "cold-welding"

of aluminium was cited as a practical application of the adhesion of metals. The adhesion between surfaces was measured recently by McFarlane and Tabor. Concluding his remarks on static friction Dr. Clayton referred to a recent paper by Parker and Hatch<sup>(13)</sup> on "The static coefficient of friction and the area of contact" and to a preprint of a paper by Parker, Farnworth and Milne on "The variation of the coefficient of static friction with the rate of application of the tangential force." He discussed Bowden and Tabor's<sup>(14)</sup> interpretation of Amontons' law and Whitehead's<sup>(15)</sup> experiments extending to low normal loads of the order of milligrams. The friction force was described as the sum of the resistance to shear of solid-to-solid contacts, drag of primary and other adsorbed films, and viscous resistance of free fluid in pockets.

Turning to kinetic friction the first experimental evidence of transfer of matter from one friction element to the other was ascribed to Bowden and Tabor, and reference was made to a paper by Strang and Burwell to be published shortly by The Institution of Mechanical Engineers showing by means of radioactive indicators, that piston-ring material is transferred to the cylinder wall during running-in, throughout the whole length of the ring travel, and in a distribution similar to that of the wear that normally occurs in an engine.

In his survey of the two characteristic types of force of friction/speed of relative motion curve, Dr. Clayton pointed out that the one exhibiting a falling, unstable section in the low speed region was the most common, applying to plain mineral oils and ordinary bearing surfaces, whereas the other, with the force of friction falling with the final decrease of speed applied to fatty oils, and to mineral oils containing a sufficient quantity of fatty oil or fatty acid. In connexion with the former type of characteristic frictional relaxation oscillations were mentioned. Passing reference was made to the effect of temperature on the friction/velocity characteristic and to the effect of roughness on friction under conditions of boundary lubrication. As far as the heat developed at rubbing contacts was concerned, Dr. Clayton thought that some of the contact temperatures found by Bowden and Ridler<sup>(16)</sup> were a surprise at the time. In this connexion Parker and Marshall's<sup>(17)</sup> work was mentioned.

Among the practical applications Dr. Clayton surveyed running-in, wear, scuffing and seizure. In his view the removal of particles of the harder metal was the result of local weaknesses initially or induced by repeated stressing in successive rubbing contacts. Shearing within the softer metal and some of it being taken away on the mating surface is visualized on account of local solid-to-solid adhesions within the tiny contact areas. At yet other contacts there would be interlocking of surface roughness with consequent shearing removal of metal; in other words, a small-scale filing process. More work is called for to elucidate the mechanism of wear. Bezer and Schnurmann<sup>(18)</sup> have

shown, in agreement with Whittaker's<sup>(19)</sup> observation, that in the region of the validity of Amontons' law the work done on abrading matter accounted for a small proportion of the frictional force. An interesting suggestion to reduce wear by the addition of phosphorus-containing materials to lubricants had been made by Beeck, Givens and Williams.<sup>(20)</sup> These phosphorus-containing materials were supposed to function by the formation of iron phosphide forming a eutectic with iron of lower melting point than iron, and flow thereby relieving local pressure without loss of material. Whilst a wear-reducing agent may be desirable in the long-term running of a mechanism, the prior use of a wear-promoting agent may be desirable to secure quick running-in of a mechanism. Dr. Clayton mentioned the engineer's definition of the terms scratching, scoring, scuffing and seizure. Scratching of the surfaces of the friction elements was regarded as a consequence of particles getting trapped between them. When the scratch was deep it became difficult to distinguish from a score such as marks the early stages of scuffing, with severe tearing of the surfaces of the solid bodies. If the load causing scuffing was temporary, and in just a few other cases, recovery to a stable condition was possible. But generally severe tearing had an accumulative effect so that conditions became worse, the driving force was exceeded, and the mechanism stalled. This is what was understood as "seizure."

Among the laboratory test machines to elucidate the behaviour of extreme-pressure lubricants Dr. Clayton thought that the I.A.E. spur gear machine seemed to have established itself most firmly for moderate duty conditions. The four-ball apparatus also seemed to enjoy some popularity although it does not give a direct imitation of gear conditions. Reference was also made to the S.A.E. machine and to a disk and straight-edge machine described by Brix.

After a reference to solid lubricants Dr. Clayton paid attention to surface treatments, particularly to phosphating by which a thin layer of mixed iron and manganese phosphates was formed at the surface.

The discussion on Dr. Clayton's address ranged over a portion of the wide field that he had covered. DR. R. SCHNURMANN (Manchester Oil Refinery Ltd.) thought that Bowden and Ridler's *experimental* results were rational and confirmed expectation. Owing to frictional heating the temperature at the actual area of contact increased with the speed for a given normal load, or with the normal load for a given speed, until the melting point of the lower-melting friction element was reached. He quoted Bowden and Ridler's experiments with gallium on steel where at a normal load of 27 g-wt the high speed of relative motion of 720 cm/sec was required to achieve a temperature rise at the *actual* area of contact by 13.2°C. There was, however, a discrepancy between Bowden and Ridler's experimental results and their calculation of "localized" high temperatures. MR. BLOK considered that the hypoid gear



difficulties were arising from very high temperatures and not from the bearing pressures. He thought that the high temperatures were caused by the super-imposition of a wiping motion to the rolling motion, and the pressures encountered with the hypoid gear were not so large as was commonly believed, since it was a question of a line contact and not of a point contact. DR. F. T. BARWELL (Department of Scientific and Industrial Research) struck a justifiably gloomy note by expressing the opinion that in view of the many factors involved in friction phenomena the prospect seemed small of arriving at an all-embracing theory of the mechanism of friction under a wide variety of operating conditions. DR. MARDLES referred in this connexion to deviations from Amontons' law even at light loads in the presence of thin adhesive films, and DR. E. TINGLE (Royal Aircraft Establishment) discussed Whitehead's results, pointing out that with some materials there was an abrupt change in the friction force/load curve at the load which caused break-up of the oxide layer.

PROFESSOR G. I. FINCH'S (Imperial College of Science and Technology, London) paper on "The structure of the boundary layer" was read by MR. R. T. SPURR (Imperial College of Science and Technology, London). Considering that in the atmosphere an oxide or sulphide layer immediately begins to form on a naked metal surface, the function of a boundary lubricant was to replace "the oxide-layer component of friction" by one of still lower value. The lubricants consisted generally of mixtures of straight-chain hydrocarbon molecules or their end-substituted derivatives which are grouped in bundles, in Professor Finch's view, like sticks in fascines. Electron diffraction patterns suggest that when grown from the melt or solution on a solid surface, these "crystals" are orientated with the chains either vertical or inclined to the surface at an angle characteristic of the compound. The thickness of such an orientated layer may be approximately 50 molecules. The heat generated at a frictional contact tends to break up the fascines. The molecules attached directly to the surface are thought to be normal or steeply inclined to it. The degree of orientation decreases from layer to layer, and the suggestion was that by the time the fourth layer was reached the orientation was practically non-existent. The maximum thickness of the boundary layer was thought to be of the order of less than 200 Å. Professor Finch further considered the possibility of the reaction of acids or esters with the oxide, sulphide or other layer to form soaps and conjectured the possibility of the relatively thick crystalline layer persisting up to the softening temperature of the soap. His paper emphasized the hypothetical nature of these views by pointing out two facts. Boundary friction was usually lower with end-substituted paraffin chains than with normal paraffinic hydrocarbons of similar chain lengths. Also, many lubricants contain, or may even consist largely of cyclic compounds. Finally, Professor Finch's paper referred to solid boundary lubricants and to the mis-

leading view that the layer-lattice crystals acted like orientated packs of playing cards. Surfaces, freshly exposed by cleavage in air, will cohere firmly on being put together again. They lose this property, however, on prolonged exposure to air or on brief immersion in water or a liquid lubricant.

MR. T. B. LANE (Thornton Research Centre), in his paper on "Boundary film lubrication failure," described a two-ball machine and its operation on the assumption of the existence of Blok's<sup>(21)</sup> "characteristic temperature for scuffing" for any given combination of lubricant and friction element. Starting from this conception an expression was derived relating "the failure temperature" to the operating variables of a test machine comprising two revolving steel balls in frictional contact. The operating variables were the rolling velocities  $v_1$  and  $v_2$ , the relative curvature of the contact, the oil temperature  $\theta_0$ , and the normal load  $W$ . Postulating that

$$\theta_0 + \Delta\theta_S + \Delta\theta_F = C$$

one has

$$\theta_0 + (1 + \alpha)\theta_F = C$$

since  $\theta_S$ , the surface temperature of the friction elements, can be expressed as a fraction of the "flash temperature"  $\theta_F$ , so that for any given oil:

$$\theta_0 + 1.99 \frac{v_1 - v_2}{\sqrt{v_1} + \sqrt{v_2}} \mu \sqrt{W}/\sqrt{R} = C$$

where  $\mu$  is the coefficient of friction,  $R$  is the relative radius of curvature, and  $C$  a constant. The view was expressed that this relationship accounts for the observed behaviour of a series of mineral oils on this two-ball apparatus. Mr. Lane checked on the basis of Howlett's<sup>(22)</sup> calculations that under his operating conditions for the two-ball machine he was outside the range of hydrodynamic lubrication.

DR. F. T. BARWELL (D.S.I.R., Lubrication Division, Mechanical Engineering Research Organization) presented his joint paper with MR. A. A. MILNE on "Lubrication with materials in the solid state." Powders of different degrees of hardness and paraffin wax were tested on the N.P.L. type machine (i.e. Bristow's<sup>(23)</sup> version of the Bowden-Leben apparatus).<sup>(24)</sup> The surface of one friction element consisted of a circular flat having an area of 0.05 in<sup>2</sup>, giving a nominal pressure at the area of contact of 40 lb-wt/in<sup>2</sup>. The lower surface was cleaned by scrubbing with caustic soda solution until, on washing with water, the surface was entirely wetted. In each case the upper surface was polished by reciprocating the lower carriage of the machine which was covered with emery paper. The coefficient of friction was determined for various solid lubricants at two speeds, at 0.01 cm/sec and at 0.4 cm/sec. Coefficients of friction between about 0.5 and 0.08 were found, with talc giving the highest value among the thirteen materials examined, and flake graphite, paraffin wax, zinc stearate and sodium stearate being at the bottom end of the list. Aluminium stearate gave a comparatively high value of 0.3 to 0.4.

Paraffin wax was further examined under continuous

running conditions using for this purpose a reciprocating machine. Here the applied load was 5 lb-wt (bearing pressure 6.7 lb-wt/in<sup>2</sup>), but in cases where a test lasted 50 hours the load was increased up to 200 lb-wt. One friction element was restrained by strips to which resistance strain gauges were attached so that the force of friction was continuously indicated by changes in resistance being registered on a cathode ray oscilloscope. The average speed over a stroke of  $4\frac{1}{2}$  in was 45 cm/sec. The average time to seizure and the average value of the coefficient of friction were determined for mild steel on mild steel with various surface treatments given to the mating area of the oscillating friction element both without and with the application of paraffin wax. The efficiency of paraffin wax as a lubricant was markedly affected by the nature of the surface treatment given to the steel plate. The phosphated surface lubricated with paraffin wax seemed the best combination, with the ground steel surface a close runner up. In the case of tin-plating the unlubricated combination gave a longer life than the lubricated, although lubrication did reduce the coefficient of friction. Force of friction/time records for applied loads of 5 lb-wt, 100 lb-wt, and 200 lb-wt show no evidence of change in the force of friction throughout the stroke for the lightest load, a square-shaped record at 100 lb-wt, whereas at 200 lb-wt the force of friction exhibited on the record a deep trough towards the middle of the stroke. Some suggestions regarding the interpretation of these phenomena were made.

In the discussion MR. E. A. SMITH (Acheson Colloids Ltd.) referred to a recent publication by Trillat<sup>(25)</sup> and DR. J. S. COURTNEY-PRATT (University of Cambridge) mentioned one of his results with multiple-beam interferometry.

In his joint paper with DR. BARWELL on "The lubrication of rough steel surfaces by a series of soaps," MR. A. A. MILNE referred to Bowden's conjecture that the boundary lubricating properties of fatty acids derive from their ability to react with some metal surfaces with the formation of metallic soaps. The frictional force of a spherically-ended friction element which was loaded against a flat plate that was moved at a constant velocity was measured. The flat plate was lapped to give a non-directional average roughness of 50  $\mu$ in, and had a hardness of 200 H<sub>D30</sub>. The spherical end had a radius of 0.5 cm and had a hardness of 850 H<sub>D30</sub>. The velocity of relative motion was varied over a range from 0.001 cm/sec to 0.6 cm/sec. Over this range there was an indication that

$$\log(\text{coefficient of friction}) \propto \log(\text{speed}) + \text{constant}$$

Experiments with oleates and stearates showed a tendency for the coefficient of friction to increase with the temperature, particularly above about 50° C. An effect of the acid radical on the coefficient of friction was apparent since at temperatures above the melting point of copper stearate there seemed to be little

difference between it and copper oleate. At lower temperatures, however, where the stearate was solid and the oleate was greasy, the stearate appeared to exhibit lower values of the coefficient of friction. With methylstearate and with a straight-chain hydrocarbon (docosane) there were discontinuities at the melting points with subsequent increases in the coefficient of friction. With stearic acid a series of measurements was made when the temperature was raised by increments of 25° C to 150° C, and then lowered. After heating to 150° C the coefficient of friction was appreciably lower both at 25° C and at 50° C than it had been before at these temperatures, due to the formation of soap. DR. BARWELL and MR. MILNE conclude from these experiments that it is the physical state of a boundary lubricant rather than its chemical composition which characterizes its quality as a lubricant. The chemical nature of the lubricant is important only in so far as it may result in a reaction with the surface of the friction elements, and may govern the rheological properties.

DR. SULLY (Boake Roberts and Co., Ltd.) drew attention to the question of the chemical purity of such materials as soaps and esters and to their acid content, and DR. BARWELL gave an assurance that the substances used were pure.

The next group of three papers commenced with MR. K. V. SHOOTER'S (University of Cambridge) contribution on "The lubrication of metals by long chain organic compounds." With a non-reactive lubricant such as a saturated hydrocarbon, boundary lubrication "broke down" at a temperature corresponding to the bulk melting point of the lubricating material. With fatty acids on reactive metals the "breakdown" occurred at a higher temperature which was said to correspond "more nearly" to the softening point or melting point of the metallic soap formed by reaction on the surface. The experiments of Tingle<sup>(26)</sup> were referred to as showing that under certain circumstances lubrication was effective only when chemical reaction occurred so that a soap film several molecular layers in thickness was formed. Further reference was made to Greenhill's<sup>(27)</sup> attempts to determine the number of Langmuir-Blodgett<sup>(28)</sup> monolayers of stearic acid, ethyl stearate, and of metal soaps necessary to give good lubrication on various metals. Shooter, in his own experiments, had adopted the Zisman<sup>(29)</sup> method of depositing monolayers of fatty acids of different chain lengths. The friction results on a Bowden-Leben apparatus indicated that lubrication seemed most effective when chemical reaction occurred with the formation of several molecular layers of soap. An attempt was made to determine the temperatures at which continuous relative motion gave way to jerky motion without changing the speed of the propelled friction element, and these temperatures were regarded as disorientation temperatures.

DR. J. W. MENTER (University of Cambridge) then gave his account of "A study of boundary lubricant



films by electron diffraction." He concluded from electron diffraction patterns that with fatty acid films on reactive metals higher disorientation temperatures would exist than for non-reactive metals such as platinum where the monolayer seemed to lose its orientation at the bulk melting point of the acid. On reactive metals both the structure and the disorientation temperature were similar to those observed with a thin film of the corresponding metal soap, and this independence for soaps of the nature of the metal substrate supported the view of a chemical reaction occurring between fatty acids and reactive metal surfaces. With non-reactive long chain polar compounds such as alcohols and esters Sanders<sup>(30)</sup> had shown that the first monolayer was strongly orientated at room temperature but lost its orientation at a temperature close to its bulk melting point. This occurred on both reactive and non-reactive metals. With some specimens of esters, however, a different observation was made. On reactive metals orientation persisted up to temperatures well above the melting point of the ester. The suggestion was that small quantities of fatty acids were present in the ester or were formed by hydrolysis at the metal surface.

DR. A. C. MOORE (University of Cambridge) presented the last of this group of papers on "The adsorption of lubricant films: a study by radioactive tracers." The technique employed was to use radioactive metal surfaces, depositing a suitable non-radioactive boundary lubricant on them, then dissolving off the monolayer and determining the metal in it. Radioactive metal foils were used and solutions of the lubricant were prepared in 20 cm<sup>3</sup> of benzene. It is claimed that reaction to the extent of 1/100th of a monolayer of organo-metallic compound can be detected. With octadecyl alcohol there was no chemical reaction with any of the metals examined. With stearic acid there was no reaction with gold or platinum, but there were reactions with zinc, cadmium and copper. If enough acid was present, the reaction did not stop with the formation of a monolayer of soap. The reaction with copper was much slower than with zinc or cadmium. Here again the reaction did not cease with the formation of a monolayer. Deliberate oxidation of the metal surfaces increased the rate of reaction. With ethyl stearate no reaction was observed with gold or platinum. A slight, but definite chemical reaction was observed with zinc, cadmium and copper. The amount of the reaction depended somehow on the specimen of the ester, and Dr. Moore suggested that small amounts of acidic impurity in the ester might be responsible for most of the reaction. Co-ordinated experiments with a Bowden-Leben apparatus suggested that effective lubrication was only obtained with those combinations of metals and lubricants which, by the radioactive results, could produce at least a monolayer of metallic soap by chemical reaction.

In the discussion of these three papers DR. W. HIRST (Associated Electrical Industries) inquired whether Dr. Menter's experiments would indicate a suspension of

the lubricant films from the peaks of the asperities of the friction elements stretching across the "valleys" rather than bedding down in them.

DR. E. W. J. MARDLES (Royal Aircraft Establishment, Farnborough) presented his joint paper with DR. E. BIELAK and DR. E. TINGLE on "The spreading behaviour and lubricating properties of mixed liquids." The rate of radial spreading of several types of binary mixtures of liquids on a number of surfaces had been correlated with their lubricating quality. The mixtures included those of various types of mineral lubricating oils, B.P. liquid paraffin with organic acids and other "polar" materials, mixtures of alcohols and esters, of lubricants with additions of fuel hydrocarbons, fatty acids and ketones. In general, where the additions to a single liquid enhanced its surface activation as indicated by the increased rate of spreading, the coefficient of friction on the same surface was reduced. The effect has been attributed to differences in the character of the surface layers consequent upon changes in the molecular composition and degree of molecular association or dissociation of the binary mixtures. The correlation was more apparent at the lighter bearing pressures, especially as might occur during the rubbing of small particles dispersed in a liquid. With increase in surface activity of the suspending mixture for the dispersed solid the specific viscosity, the rate of sedimentation and the volume of sediment of the suspension decrease. In the experiments the surfaces of the polished flats were washed and polished under water, then rinsed with a jet of hot distilled water and finally polished with soft lint. A known volume of liquid was delivered to the centre of the horizontal plate and its rate of spread was determined. In the friction tests a period was allowed to elapse for the oil to wet the surface thoroughly and the flat to sink into the pool of lubricant.

DR. R. SCHNURMANN (Manchester Oil Refinery Ltd.) discussed "The electrostatic component of the force of friction." When a metallic frictional contact was made to perform relaxation oscillations each jerk was accompanied by a galvanometer deflexion when, for instance, a ballistic galvanometer was connected to the two metallic friction elements.<sup>(31)</sup> The thesis was that each of these galvanometer kicks indicated an electric discharge current which arose when the dielectric breakdown field strength of the dielectric material sandwiched between the metallic friction elements had been reached. On this view the cycles of charging and discharging must follow each other in increasingly rapid succession with increasing rate of propulsion while for a given lubricant and given operating conditions (apart from the variation of average speed of relative motion) the magnitude of the galvanometer deflexion must remain unaltered. At a rate of propulsion at which the discharges follow each other in quick succession a ballistic galvanometer was unable to resolve them, whereas they could be shown on the screen of a cathode ray oscilloscope. In agreement with the electrostatic interpretation, oscillations

whose amplitude was independent of the speed of relative motion of the friction elements, could be observed even at rates of propulsion at which jerky motion had given way to continuous motion, i.e. in the stable region of the force of friction/velocity characteristic.

With the assistance of Mr. H. J. BEZER and Mr. W. J. MORRIS the speaker showed some simple experiments illustrating the universal occurrence of contact electrification without requiring any hard rubbing. The electrification of frictional contacts was illustrated with a metallic friction element loaded against a rotating steel disk when at a linear speed of several hundred centimetres per second rapid oscillations were demonstrated on the screen of a cathode ray oscilloscope. Breathing on the steel plate tended to annihilate the oscillations. On the other hand, pouring a dielectric material on to the friction track appreciably increased the amplitudes of the oscillations as compared with those observed under "dry" conditions. On replacing the revolving steel disk by a glass plate luminous spots at the frictional contact could be observed through the glass plate at temperatures below  $400^{\circ}\text{C}$  provided that the metallic friction element which was loaded against the revolving glass disk was made to tear a track to prevent smearing of metal on to the glass plate. It was shown that rubbing the glass plate with cotton wool increased the amplitude of the oscillations, whereas breathing on the glass plate annihilated the oscillations. Furthermore, a metallic probe set up behind the frictional contact and about 1 mm above the glass disk picked up static charges from the frictional contact.

One of those present wished to know for frictional contacts the electrostatic charge density, which was estimated at  $15\text{ e.s.u./cm}^2$  <sup>(32)</sup> Mr. BLOK wondered whether the electrostatic interpretation of jerky motion was an alternative to the non-linear falling section of the force of friction-velocity characteristic. Dr. BARWELL asked whether the thermal capacity of the thermocouple junction with which the appearance of luminous spots at temperatures below  $400^{\circ}\text{C}$  had been found might be responsible for the readings not being higher. This led to a brief discussion of what would appear to be the fundamental *impossibility* of generating localized high temperatures on a metal surface owing to the notorious high thermal conductivity of metals. Dr. Barwell was informed that an estimate of 10% had been made as the proportion of the force of friction which the electrostatic component assumed. Dr. MENTER thought that if the luminous spots were indications of electric discharges the nature of the gas atmosphere should affect the spectrum of the scintillations. One speaker said that the dielectric breakdown field strength of very thin films (smaller than  $10^{-5}\text{ cm}$  in thickness) could be expected to be higher than the dielectric breakdown field strength of the same materials in bulk. Mr. J. E. C. STRINGER (Research Dept., Vickers-Armstrongs Ltd.) suggested that with the avail-

able lecture demonstration apparatus an experiment with castor oil be carried out on the spot. Mr. Stringer provided a sample of castor oil and the experiment was done of showing much reduced amplitudes of the oscillations as compared with a mineral oil. Dr. COURTNEY-PRATT reiterated the arguments contained in the literature concerning localized high temperatures and he added that he had been unable to confirm that the electrostatic component formed an appreciable proportion of the force of friction. Under his experimental conditions it was nearer 0.05% of the force of friction.

The second paper by PROFESSOR G. I. FINCH and Mr. R. T. SPURR on "Surface welding and the extreme pressure lubricant," was presented by Mr. Spurr. The surface of a metallic single crystal can be made atomically smooth by appropriate anodic polishing. By suitable precautions the surface oxide film can be restricted to a few molecules in thickness. Two such single crystal surfaces do not adhere appreciably if pressed together at room temperature provided that the pressure is not sufficient to produce extensive plastic flow. The oxide film, thin though it may be, prevents direct metal-to-metal contact. On the other hand, if extensive flow took place, the authors considered that the increase in surface at the interface ruptures the oxide film and "cold welding" occurs. Consequently, the protection afforded by the oxide layer in this case was determined by the rigidity of the underlying metal rather than by the nature of the oxide. The idea was developed in this paper that during relative motion the actual area of contact increases rapidly for soft ductile metals which therefore tend to "weld" more easily than harder elastic metals. Copper single crystals pressed together gently with the fingers "weld" so strongly, after relative motion over only a fraction of a millimetre, that they are difficult to pull apart with the fingers. When different metals are in frictional contact, the softer metal may be appreciably deformed and suffer surface damage, and its oxide film may be ruptured to a considerable extent while the harder metal is relatively undamaged. The paper expressed the view that in metal pairs capable of forming low melting point eutectics it appeared that the actual interface of the "weld" has the eutectic composition. Thus, if pieces of tin and lead form the elements of a frictional contact and become so "cold welded," the blocks fall apart at precisely the melting point of the eutectic as the result of heating in vacuo. Lead and bismuth form a similar combination. Professor Finch asserts that oxide films give very effective protection against this "cold welding." If electro-polished copper specimens were exposed to the air for some time they became increasingly difficult to "weld," but if they were treated with potassium cyanide to dissolve the oxide they once again "welded" easily. The thicker the amorphous oxide film, the more protection it afforded. Conditions were different when the oxide film was not amorphous. If a copper surface was oxidized to form a crystalline layer of cuprous oxide



sufficiently thick to give interference colours, the surfaces "welded" very easily.

With other chemical films it was found that in general an amorphous film was more effective in reducing inter-metallic "welding" than the corresponding crystalline film. The properties of the sulphide film on copper were of particular interest. Crystalline films of about 1 000 Å thickness prevented "welding" and gave a low coefficient of friction under moderate pressure and low rates of relative motion. Plucking and shearing occurred within the sulphide films, and for this to happen they must be thicker than about 500 Å.

The protection afforded by extreme pressure additives was of a different nature. The surfaces of the friction elements were of hard material and the chemical films may be only 100 Å thick. Under very severe conditions high temperatures may be developed. Before the temperature at the actual area of contact became high enough to soften the underlying metal and thus rob the oxide layer of its support, the additive reacted with the oxide to form a layer of a compound such as a chloride or sulphide which was more fusible than the metal. Professor Finch considered that if the "local temperature" should rise sufficiently to melt this compound, some of the molten layer would smear over the surface and act as a temporary lubricant.

DR. C. H. M. JENKINS (National Physical Laboratory, Teddington) presented his joint paper with DR. CLAYTON on "Physical changes in rubbing surfaces on scuffing." While this work was a continuation of an established National Physical Laboratory programme of investigation of the scuffing of surfaces, practical difficulties experienced in running-in a particular type of new aero-engine gave it impetus in the early stages of the last war. Scuffing of the cast-iron piston rings occurred with mineral oils and satisfactory freedom from it could be obtained only with castor oil. Experiments were made in which portions of piston ring in the form of segments were substituted for the three stationary balls of the four-ball apparatus. Scuffing of piston rings occurred at about 250 kg-wt with mineral oil. Scuffing of steel balls occurred at lower loads of about 60 kg-wt. The coefficient of friction during scuffing was never so high with cast-iron rings as with steel balls. Microscopic examination revealed a white constituent in the structure of the wear impressions. The surface structures of three typical piston rings after running-in in aero-engines showed that the surface of one which had "seized" in the engine was almost covered by the layer of white constituent, that the surface of an unsatisfactorily run-in ring had considerable amounts of white constituent in it, and that no white constituent was present in the surface of a ring which had run-in satisfactorily using castor oil. It appeared that the white constituent could be produced freely with mineral oil when high loads caused scuffing or when the "scuffing load" was approached. It could also develop in the case of prolonged running times at lower load. Experi-

ments were made to try and identify the white constituent. Etching reagents had shown it not to be martensite, but it etched in hot sodium picrate solutions in a similar manner to iron carbide. The amount of white constituent was larger with the high carbon content rings than with the 1.2% carbon balls. Under the low load conditions no white constituent was formed with castor oil. It appeared that the mineral oil did not provide a source of carbon as good as graphite in the rings even at the high temperatures of rubbing. In an experiment with a carburized pure iron no white constituent was formed in a non-scuffing run with plain mineral oil. However, with a concentrated colloidal graphite mixture instead of plain oil, scuffing occurred at the same load and the test had to be stopped quickly. Both the test pieces and the ball showed a heavy layer of white constituent with chatter marks in the white layer. A quenching test suggested that as a result of extreme quenching speeds unusual conditions arise in carbon steels in the austenitic region at high temperatures. The quenching products are resistant to etching in a similar manner to the white constituent. It appears that at the temperature of formation the white constituent consists of the two phases cementite and austenite. Normally the austenite would form martensite, but for some reason it forms a constituent similar to  $\alpha$ -iron, which would be supersaturated with carbon.

Wear tests indicated that "the scuffing load" for castor oil for a ball running on three piston ring segments was about 500 kg-wt, compared with 250 kg-wt for plain mineral oil. There was no difference when the free fatty acid was carefully neutralized and the soap removed. The addition of 1% ricinoleic acid to plain mineral oil had no effect on its "scuffing load." Several additives were tried including tricresyl phosphate, but the only one giving an increase of "scuffing load" with piston rings was 1% of thio-ether. "The scuffing-load" was as high as with castor oil. The wear at loads just below the scuffing range was greater with the thio-ether than with the plain mineral oil. With wear-reducing additives similar results to those of Beeck, Givens and Williams<sup>(16)</sup> were obtained. At low loads their conclusions on the benefits in wear reduction by phosphorus and arsenic containing additives were confirmed. Sulphur and chlorine compounds of the extreme-pressure type were also found to have wear-reducing properties. 0.2% colloidal graphite was not effective in this respect. Castor oil was an effective wear reducer when tested with a steel ball on steel balls. With a steel ball on piston ring segments there was the same wear of the segments as with the plain mineral oil, but the wear on the ball was much less. The same result was obtained with castor oil without free fatty acid, and with 1% ricinoleic acid in mineral oil. With 1½% oleic acid in mineral oil the wear on both rings and balls was reduced to a small value. Experiments were made with two chemical surface pre-treatments. One consisted in etching with nitric acid intended to remove superficial ferrite. There

was no improvement in "scuffing load," using mineral oil, and the wear of piston ring segments and ball at low loads was only slightly better than with plain rings. The other treatment was phosphating. This resulted in a striking increase of "scuffing load" with plain mineral oil leading to the top load of the apparatus (750 kg-wt). The engine experience showed phosphated rings to be very much superior to those etched with nitric acid.

The discussion of the last two papers was taken together. DR. PARKER strongly emphasized the need for an explicit definition of the terminology particularly of such terms as "welding" when used in connexion with friction, and DR. CLAYTON supported this proposal, suggesting that it might be desirable to set up a committee for the purpose of trying to standardize the terminology. DR. J. S. MCFARLANE (Imperial Chemical Industries Ltd.) and MR. SPURR discussed the forces required for the separation of soft metals with regard to the possible part played by the oxide film and to the area of contact. Dr. McFarlane referred to his experiments using a pendulum type of apparatus.<sup>(33)</sup> DR. E. M. CROOK (Associated Electrical Industries Ltd., Aldermaston) pointed out differences found in the forces needed for the separation of soft metals, such as indium, depending on whether static or dynamic loading had been applied. MR. STRINGER raised the point of the effectiveness of sulphur-containing compounds as extreme-pressure additives, and of the development in oil refining operations accounting for lower sulphur contents of present-day lubricants, since he wondered whether it was the quality of cast iron or the quality of the lubricants that had changed during the past thirty years and was responsible for the change in the behaviour of cast-iron bearings. DR. CLAYTON's opinion was that the cast iron had changed and that its lubricating quality was better thirty years ago, whereas its mechanical strength had improved by 1940. With reference to the sulphur content of solvent refined oils MR. E. A. EVANS thought that there were one or two or at the most four sulphur compounds which could be regarded as suitable "extreme-pressure" additives to a mineral oil.

MR. E. RABINOWICZ (University of Cambridge) delivered his paper on "An investigation of surface damage using radioactive metals." He showed the well-known taper section photomicrograph where a few small splinters of copper have been picked up by a steel surface. This pick-up may be very small in the presence of a good boundary lubricant. Under these conditions very sensitive methods for detecting the pick-up must be used, and of these the most effective was the radioactive method. In his experiments a radioactive metal was rubbed with its hemispherical end over the surface of a non-radioactive metal. Any pick-up of metal from the curved friction element caused the blackening of a photographic film placed in contact with the second surface. In favourable cases the method detects pick-up down to  $10^{-12}$  g. With a good boundary lubricant the

pick-up may be reduced by a factor of as much as 10 000 as compared with "dry" conditions. As an example, palmitic acid as a lubricant may give a pick-up of  $10^{-7}$  g/cm<sup>2</sup> under conditions under which without a lubricant the pick-up was  $1.4 \times 10^{-4}$  g/cm<sup>2</sup>. If lubricated surfaces are heated, the lubrication ceased to be effective at a typical "breakdown temperature." This transition was marked by an appreciable increase in the force of friction, and the radioactive measurements showed that this was accompanied, in general, by a corresponding rise in the pick-up. There was no simple relationship between the force of friction and pick-up. Boundary lubricants giving the same coefficients of friction may give widely differing amounts of metallic transfer.

MR. P. GRODZINSKI'S (Industrial Distributors (Sales) Ltd., Diamond Research Department, London) paper on "The influence of lubricants on friction, wear and abrasion" concluded the programme of the Conference. He said that lubricants in abrasion processes serve to remove the abrasion products from the frictional contact and to enhance the abrasive action. The author's earlier tests had suggested a coefficient of friction of 0.3 for diamond grinding with the ordinary grades of powder, whereas more recent investigations with a very uniform powder of up to  $0.2 \mu$  showed a coefficient of friction of about 0.1. It may be possible, though it is not certain, that lubricants which do not behave well in bearing tests are suitable fluids for abrasion processes. In the machining of hard materials Mr. Grodzinski distinguishes six essential factors:

(1) the grinding speed, (2) auxiliary (lapping) movement, (3) the pressure, (4) the grinding and polishing grain (of suitable and uniform size), (5) lubrication of the stock and grinding wheel, (6) type of "bearer" or "carrier" material for "loose" and "bonded" grain.

The effect of the lubricant depends on the amount and the nature of lubricant supplied. In diamond grinding one distinguishes between wet and dry preparation of the cast-iron disk, and Mr. Grodzinski referred to Kruehl's<sup>(34)</sup> experiments with metal wheels into which diamond dust had been rolled. Starting under dry conditions the rate of abrasion at first increased, and on applying a lubricant it slowly decreased with increasing supply of lubricant (water). The increased capacity in wet grinding is explained by Kruehl in terms of two possible phenomena. In dry grinding the swarf is not always fully removed, and the portion which sticks to the surface of the stock protects it against further attack. The second phenomenon is the greater stickiness owing to the limited amount of lubricant. In grinding operations the action of the same fluid depends upon the type of abrasive, and also on the type of lap. Some years ago A. F. Jones suggested the use of cyclohexanol as a carrier of diamond powder in the drilling of diamond dies. Whereas for comparable drilling times powder made up with olive oil or with lanoline enlarged a die by 0.00025 in, the use of cyclohexanol enlarged it by



0.00300 in. Lunt, Burns and Dean obtained a similar improvement with a mixture of about 5 parts of carbon tetrachloride and 1 part of diamond powder. Cyclohexanol solidifies below 300° C. Therefore, only one part of the paste which does work during the grinding operation became fluid. If centrifugal action threw off part of the fluid, it solidified and was thus not lost. In the centreless grinding of sapphire on copper wheels prepared with diamond the usual water lubrication was replaced by a thin oil film with the result that a much better surface finish was obtained. v. Engelhardt<sup>(35)</sup> found that the differences in the behaviour of various fluids could be explained by the surface energy and concluded that 1/1000 of the work done corresponded to the real surface energy, and the bulk of the energy was used for the breaking up of particles.

## NOMENCLATURE

The phenomena encountered in work on friction and lubrication in common with other phenomena involving surfaces, are of a complex nature. For this reason great care is required in the analysis of experimental results and in the choice of terms borrowed from the field of engineering lest by continual usage they might be regarded as having a distinct physical meaning. This applies to an indiscriminate usage of such terms as "welding" irrespective of whether there is or is not any interdiffusion of crystal grains,<sup>(36)</sup> to the application of terms such as "breakdown load" and "breakdown temperature" even in cases where a region of continuous transition is apparent rather than a discontinuous change. There is a need for discriminating between the relative motion of solid bodies without resultant macroscopic damage of the surfaces (the term "sliding" has been suggested in the literature for these phenomena), and with severe destruction of the surface quality ("tearing"). If the Conference has the result of leading to a clearly defined use of common engineering terms in the physical literature it may be regarded as having made one important contribution towards progress in the fields of friction and lubrication. Incidentally the Conference also gave a good idea of the scope of some of the modern tools of physics in their application to the study of friction and lubrication.

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R. SCHNURMANN

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The papers and the discussion on them will be published in full early next year in a special issue of this Journal, in place of in the Institute's "Physics in Industry" series as previously announced.

## ORIGINAL CONTRIBUTIONS

## Studies of the viscosity and sedimentation of suspensions

## Part 1.—The viscosity of suspension of spherical particles

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After a brief review of the relative viscosity/volume concentration relationships which have been suggested for suspensions of smooth spheres, the results of new experimental work on rigidly defined systems of methyl-methacrylate spheres suspended in aqueous solutions are described and discussed. It is shown that, although their relative viscosity is independent both of the viscosity of the suspending liquid and the absolute size of the spheres at a given concentration, it is a function of their size distribution, decreasing with increasing size range to a constant value. The proportionality constant between specific viscosity and volume concentration at infinite dilution varies from about 4.0 for a size ratio of 1:1 to 1.9 for ratios exceeding 3:1. When the suspensions consist of dielectric particles in a non-polar liquid, electrostatic charging of the particles may affect the measured viscosity.

Any physical interpretation of viscosity/volume concentration measurements made on non-flocculating suspensions of discrete, solid particles is generally based upon comparisons with simple systems of geometrically shaped bodies, modified to fit the observed results. The most widely used relationship is that derived by Einstein<sup>(1)</sup> for smooth, rigid, inertia-less, spherical particles at infinite dilution, according to which the relative viscosity  $\eta_r$  is given by

$$\eta_r = \frac{\eta}{\eta_0} = 1 + kc$$

where  $\eta$  = viscosity of the suspension,  $\eta_0$  = viscosity of the pure liquid,  $c$  = volume concentration of the suspended particles,  $k$  = constant = 2.5.

Simha,<sup>(2)</sup> Jeffery,<sup>(3)</sup> Burghers,<sup>(4)</sup> and Vand<sup>(5)</sup> have all derived similar relationships, and the value of 4.5 obtained for  $k$  by Hatschek<sup>(6)</sup> is not considered to be reliable.<sup>(7)</sup> Philippoff has pointed out<sup>(8)</sup> that the experimental verification of the value of  $k$  rests almost entirely upon the work of Eirich, Bunzl and Margaretha;<sup>(9)</sup> according to an analysis of their results made by Mark,<sup>(10)</sup>  $k$  may vary from 1.5 to 3.0 depending upon the suspension and viscometer used. Other workers<sup>(11-20)</sup> have obtained values varying from 1.9 to 4.7, but their suspensions or procedures are, in many cases, suspect. On the other hand, there is general agreement that the absolute size of the particles and the viscosity of the suspending liquid have little effect upon the relative viscosity.

For higher concentrations of spherical particles, theoretical relationships have been established by Guth and Simha<sup>(21)</sup> and Vand,<sup>(5)</sup> while empirical equations have been suggested by Eirich, Bunzl and Margaretha,<sup>(9)</sup> Kunitz,<sup>(22)</sup> de Bruijn<sup>(23)</sup> and Robinson.<sup>(24)</sup> Experiments have been made on suspensions of a wide variety of materials<sup>(9, 11-20, 22-24)</sup> whose sphericity, hardness, surface smoothness or chemical inactivity can, in many cases, be questioned. An appraisal of these experiments has been made recently<sup>(25)</sup> and it fails to show the absolute validity of any one of the published theoretical

or empirical relationships for the viscosity of a suspension of smooth, spherical particles.

Some new experiments, made on suspensions of spheres, are described below.

## AQUEOUS SUSPENDING LIQUID

*The Suspensions.* The most satisfactory model suspensions used by earlier workers have consisted of glass spheres suspended in solutions of the same density. The chief objection to this type of suspension is in the necessity of using either organic liquids, which may cause flocculation of the spheres, or unstable aqueous solutions, which are difficult to handle, as the suspending liquid. In the present experiments, suspensions of methyl-methacrylate polymer spheres in an aqueous solution of lead nitrate and glycerol of viscosity about 6 centipoises were used and found to remain stable and unflocculated even after a period of a week. These particles have the advantages of low density, small water absorption and almost perfect sphericity. Four samples, which were produced by dry and wet sieving and elutriation, are detailed in Table 1; photomicrographs of samples (a) and (d) are shown in Figs. 1 and 2 respectively.

Table 1. Spherical powders used in viscosity measurements

Sample	Size range Microns	Size ratio
		Largest particle Smallest particle
(a)	152-177*	1.17
(b)	147-208*	1.414
(c)	76-208*	2.74
(d)	27.5-49.5†	1.8

\* Flat-topped size-distribution.

† Normal size-distribution, 83.5% lying within size-range stated.

*Measurement of Viscosity.* A rising-sphere type viscometer was selected because, by making the ratio of sphere to suspension-tube diameter sufficiently large, the effect of the slip of particles at the shearing surfaces (see Vand<sup>(5)</sup>) becomes negligibly small. The tube was 180 mm tall and 30 mm in diameter, and the spheres



of approximately 1.4 mm diameter, were released from two different depths, their time taken in reaching the suspension surface being measured in each case. The spheres were of polystyrene and methyl-methacrylate mixtures, pigmented to facilitate identification; their characteristics are summarized in Table 2. The measurements were made in a thermostat maintained at  $25^{\circ}\text{C} \pm 0.05^{\circ}$  on suspensions of each of the four powders, the concentration being increased from zero to 30% by volume, in increments not exceeding 5%.

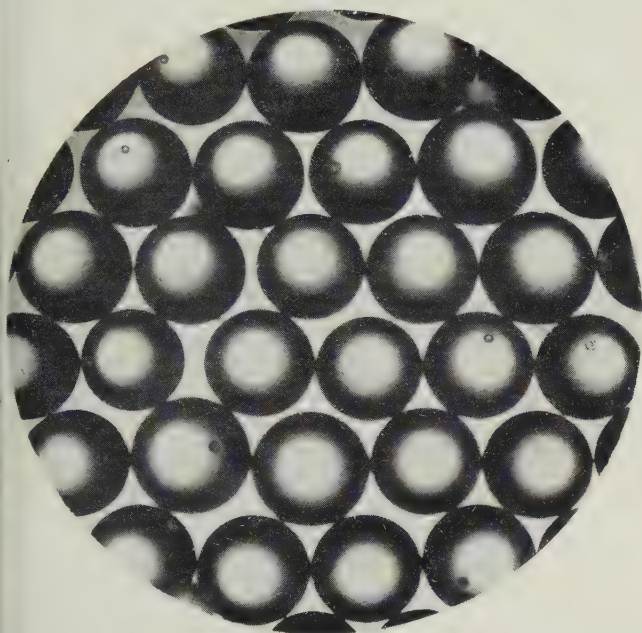


Fig. 1. Photomicrograph of methacrylate spheres, sample (a)

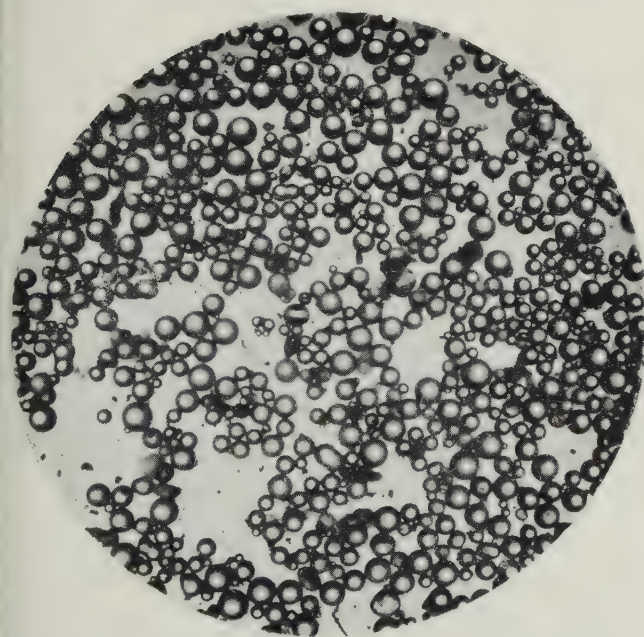


Fig. 2. Photomicrograph of methacrylate spheres, sample (d)

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Table 2. Dimensions of rising spheres

Colour	Density g/ml	Diameter cm
Pink	1.1783	0.1402
Blue	1.1720	0.1415
Cream	1.1525	0.1491

**Results.** Each sphere velocity was corrected for the presence of the tube walls by the Faxen correction (see Barr<sup>(26)</sup>). The maximum Reynolds numbers reached by the spheres were 0.5, 0.8 and 1.8 for the pink, blue and cream spheres respectively. Up to a Reynolds number of 0.1 the viscosity was calculated from Stokes's law, but above this value an empirical relationship obtained by Davies,<sup>(27)</sup> which was found to be accurate to within 1% for Reynolds numbers up to 0.8 and to within 2.25% up to 1.8, was used, small additional corrections being applied at the higher sphere-velocities to correct for these slight discrepancies. It is estimated that the resulting viscosity measurements were accurate to within  $\pm 2\%$ . Fig. 3 shows the variation of relative viscosity with volume concentration for the spherical powders. Extrapolation of the relative viscosity/size ratio relationships shown in Fig. 4 yields the fifth curve in Fig. 3.

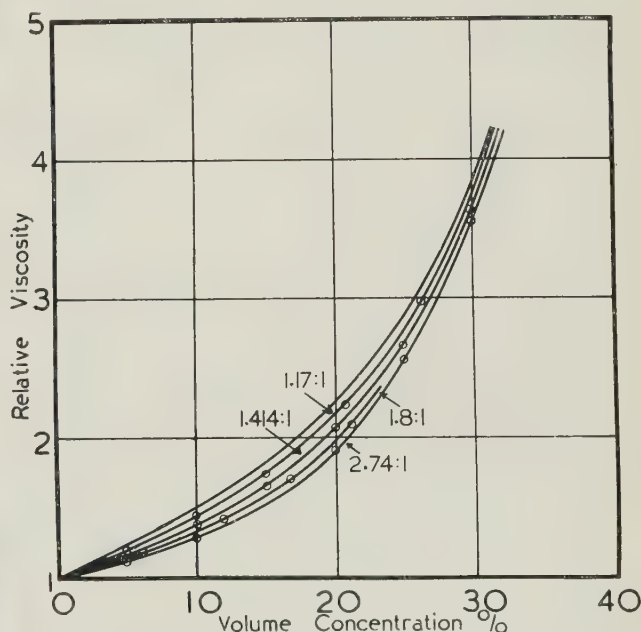


Fig. 3. Relationship between relative viscosity and volume concentration of suspensions of spheres of various size ratios

**Discussion.** It is concluded that the relative viscosity of a suspension of hard, smooth, non-interacting spheres at a given concentration when measured in a rising-sphere viscometer is:—

1. Practically independent of the rate of shear up to volume concentrations of 30%.
2. Independent of the absolute size of the spheres.
3. Independent of the viscosity of the suspending liquid.

4. Dependent upon the size distribution of the spheres, decreasing with increasing size-range to a constant value.

The Einstein constant  $k$  varies from about 4.0 for an extrapolated size-ratio of 1:1 to 1.9 for size ratios exceeding 3:1. Einstein's theoretical value of 2.5 is given for a size ratio of about 1.5:1.

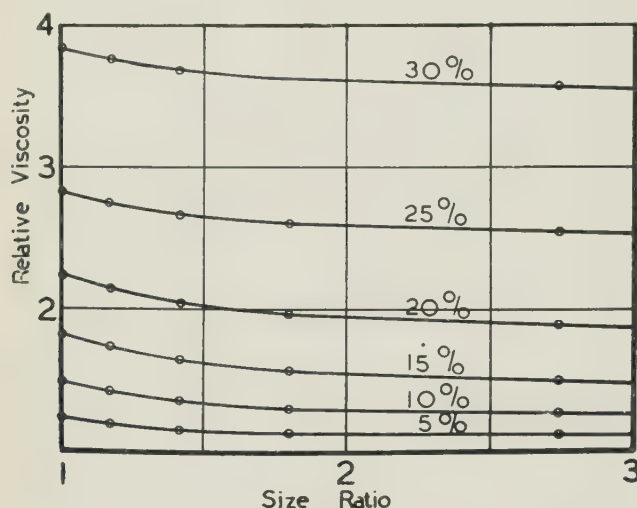


Fig. 4. Variation of relative viscosity with size ratio of suspensions of spheres at various volume concentrations

Comparison with the results of other workers is difficult because the size distributions of their solid, dispersed phases are not generally given. Moreover, there is no conclusive experimental proof that different basic types of viscometers will give identical viscosities on the same suspension. Even at low concentrations the results of Eirich, Bunzl and Margaretha<sup>(9)</sup> show appreciable variations from one instrument to another, the discrepancies being explained away in terms of inertial forces. In Table 3 a comparison is made with the results of Vand,<sup>(16)</sup> Eirich,<sup>(9)</sup> and Eilers,<sup>(20)</sup> who used aqueous suspending-liquids, and in Table 4 with those of Broughton and Windebank<sup>(19)</sup> and Robinson<sup>(24)</sup> who used non-polar liquids. The former are in better agreement with the results now reported than the latter,

Table 3. Comparison of results (aqueous suspensions)

Volume Concentration (%)	Vand*	Eirich†	Relative viscosity Size Ratio§ 1.6:1	Eilers‡	Size Ratio§ 3:1
0	1.00	1.00	1.00	1.00	1.00
5	1.145	1.132	1.125	1.10	1.10
10	1.34	—	1.32	1.25	1.25
15	1.62	—	1.59	1.49	1.47
20	2.02	—	1.99	1.80	1.84
25	2.63	—	2.63	2.16	2.53
30	3.64	—	3.65	2.59	3.53

\* Ostwald capillary tubes. Glass spheres 100–160  $\mu$  in aqueous solution of zinc iodide and glycerol, 80 cp.

† Falling-sphere viscometer. Glass spheres 100–160  $\mu$  in saturated solution of mercuric iodide in concentrated nitric acid, 20 cp.

‡ Vogel-Ossag capillary viscometer. High viscosity bitumen 1.6–8.1  $\mu$  in aqueous potassium soap. Viscosity not stated.

§ From Fig. 4.

Table 4. Comparison of results (non-polar suspensions)

Volume concentration (%)	Relative viscosity Broughton*	Robinson†	Size Ratio§ 2:1
0	1.00	1.00	1.00
5	1.15	1.16	1.12
10	1.35	1.36	1.30
15	1.61	1.62	1.55
20	1.94	1.98	1.95

\* MacMichael rotating-cylinder viscometer. Glass spheres 37–74, 74–147, 147–295  $\mu$  in mixture of acetylene tetrachloride and monobromobenzene. Viscosity not stated.

† Buchdahl rotating-cylinder viscometer. Glass spheres 10–20  $\mu$  in S.A.E. oils Nos. 30 and 50 and in castor oil.

§ From Fig. 4.

particularly at concentrations up to 20%. Thus for smooth spheres of size ratio about 1.5:1 Einstein's and Vand's theories are confirmed, the former for dilute suspensions and the latter up to volume concentrations of 30%. Vand's theory, however, assumes that the spheres are all of equal diameter, and the results now reported yield a different relationship under such conditions. The most important difference is that at infinite dilution they give a value of  $k$  of about 4.0.

#### NON-POLAR SUSPENDING LIQUIDS

*The Suspensions.* When dielectric particles are suspended in a non-polar liquid, the physical properties of the resulting suspension may be affected. Experiments were made on methyl-methacrylate polymer spheres suspended in two re-distilled oils of differing viscosities, but refractive indices made equal to that of the suspended particles by the addition of small quantities of naphthalene. The particles were of two sizes, 88–104 and 147–175  $\mu$ . Details of the suspensions are given in Table 5.

Table 5. Details of particles and oils used

	Density g/ml	Refractive index	Viscosity cp
Particles .. ..	1.1893	1.495	—
Suspending oil (1) ..	0.8775	1.495	5.43
Suspending oil (2) ..	0.8870	1.495	89.28

*Measurement of Viscosity.* Since the density of the oils was less than that of the particles, the suspensions slowly sedimented. A falling-sphere type viscometer was used for the viscosity measurements, the falling velocity of the spheres being measured with the aid of a short-focal-length cathetometer in an area well above the settled particles. The observation cells were flat-bottomed, cylindrical, glass vessels, 180 mm tall and 30 mm in internal diameter, immersed in a thermostat maintained at  $25^\circ\text{C} \pm 0.05^\circ$ . The majority of the seven sets of spheres, each having a different density, were of polyethylene weighted with zinc oxide to the required density and pigmented to facilitate identification. Higher density spheres were produced from polystyrene and acrylic plastics. The suspensions were well mixed initially to ensure complete homogeneity; immediately on the cessation of stirring, a stop watch was started and spheres of a particular density introduced



successively down the axis of the viscometer by means of a drop-tube. Measurements were made at volume concentrations from 14 to 35% in increments of about 5%.

**Results.** The falling velocities of the spheres were corrected for the presence of the cylinder walls by the Faxen correction (see Barr<sup>(26)</sup>) and for the effective density of an unstable suspension to a falling body being slightly different from its measured value, by a correction due to Hirst.<sup>(28)</sup> The viscosities were then calculated from Stokes's law.

**Discussion.** The individual values of relative viscosity at a given volume concentration varied considerably from measurement to measurement, but there was a general tendency for it to increase with time from an initial value rather less than that for a similar suspension in an aqueous solution to a value considerably exceeding it. The suspensions exhibited, in fact, thixotropy as defined by Pryce-Jones.<sup>(29)</sup> The rate of increase of the relative viscosity with time at any volume concentration was greater for the larger particles and the relative increase in viscosity was greatest at the lowest concentrations.

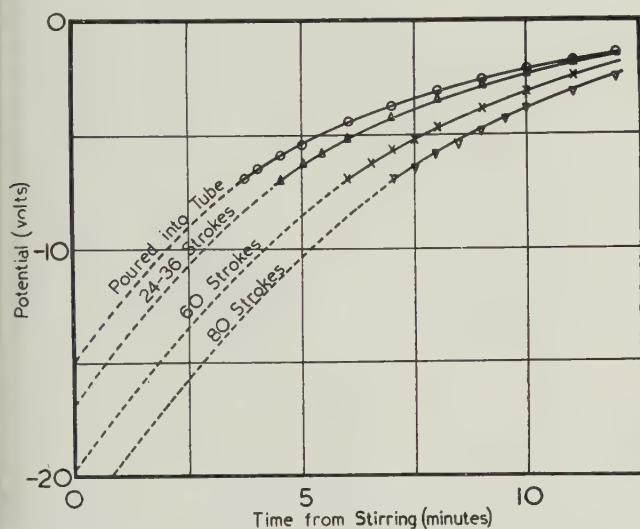


Fig. 5. Variation with time after stirring of the potential difference in a suspension of settling spheres

The accuracy of measurements which were made of the yield-values of the suspensions, using the method of Williams and Fulmer,<sup>(30)</sup> was limited by the considerable fluctuations in the rate of fall of the spheres. In the low-viscosity oil, a very small positive yield-value was detected, but the high-viscosity oil gave a negative value to which no physical significance could be ascribed. It thus appeared that forces additional to the pure hydrodynamic forces associated with the shearing of the suspensions were being measured. The sedimentation rates, however, were the same as in aqueous solutions of similar viscosity and density and agreed closely with

the results obtained by Steinour<sup>(31)</sup> for the sedimentation of spherical particles. Moreover, the sedimentation volumes were almost identical to those obtained in aqueous solutions and no flocculation was detected at any time. Sedimentation-potential experiments were made in a cylindrical settling tube, earthed on the outside and fitted with two platinum electrodes spaced vertically 110 mm apart. The potential developed between the electrodes immediately after stirring was so large that the valve voltmeter was unable to record it satisfactorily, but it died away exponentially with time. Generally, the greater the initial agitation, the larger was the potential developed in the suspension, although it was difficult to reproduce the results exactly. Earthing the brass stirrer or replacing it by a glass one made no appreciable change in the results. Fig. 5 gives some typical decay curves for 147–175  $\mu$  particles suspended in the low-viscosity oil at 15.9% volume concentration. No detectable potential was found when the oil was replaced by an aqueous solution of glycerol and lead nitrate.

It is difficult to believe that the potentials measured resulted from zeta potentials at the surfaces of the spheres since such charges, which are usually associated with electrolytes, arise spontaneously and should not decay with time. Assuming that sufficient charge could be built up on the particles from the adsorption of traces

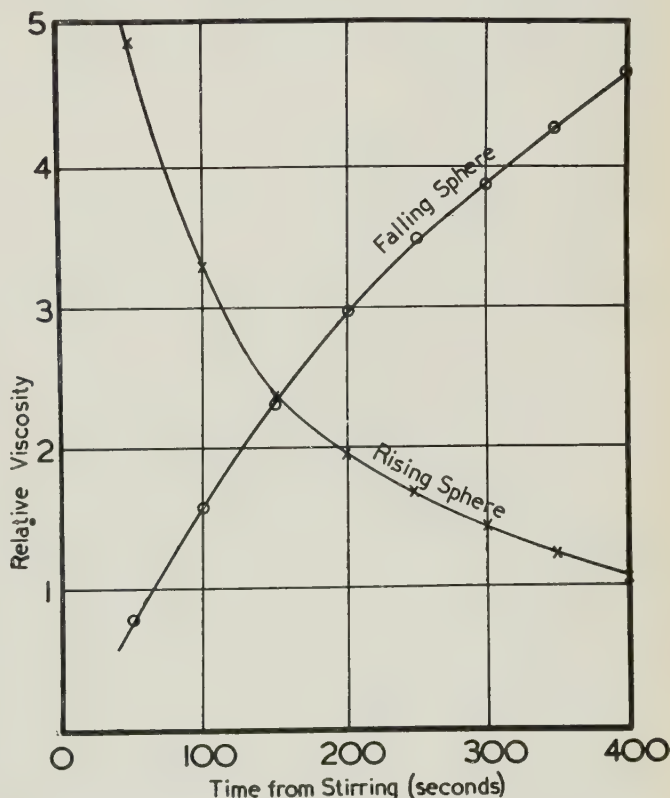


Fig. 6. Variation of apparent relative-viscosity with time as calculated from rising and falling spheres in the same suspension

of ions (as Alexander and Johnson<sup>(32)</sup> suggest), the potential measured between two points down the settling suspension should remain constant until the top electrode is uncovered.

In fact, the potential started to fall as soon as stirring ceased. Further, the initial potential was influenced by the amount of agitation given to the suspension after it had been completely dispersed, and it must, in consequence, be presumed that the charge on the particles varied from one experiment to another. It is more probable that when the suspension was stirred, the suspended particles became charged electrostatically. When stirring ceased, the charge leaked away continuously through the suspending medium.

If the electrostatic field acted in such a direction that it caused the velocity of fall of a sphere to decrease continuously with time, it should cause the velocity of rise of a sphere of similar material to increase continuously with time. This was tested by releasing weighted polyethylene spheres from the top and bottom of the viscometer tube. The apparent relative-viscosity of the suspension was then calculated for each sphere, after wall and density corrections had been applied to the results. Fig. 6 shows the results obtained for a suspension of 147–175  $\mu$  particles at 21.5% volume concentration suspended in the high-viscosity oil. The point of inter-section of the two curves, which should correspond to the true relative-viscosity of the suspension, gives a value of 2.33. This compares well with 2.29 obtained for a suspension of similar particles in an aqueous solution.

#### CONCLUSIONS

It appears that many of the discrepancies between the results of various workers may be due to the size distributions of the particles used. It is of particular interest to note that the experimental confirmation of Einstein's law rests almost entirely upon the work of Eirich, Bunzl and Margaretha, who used glass spheres of size ratio 1.6 : 1, fungal spores of ratio 1.42 : 1 and yeast organisms of unspecified size range. By adjusting the size distribution of the spheres, values of  $k$  varying from 1.9 to 4.0 are possible. It should not be assumed, however, that those results can be applied unreservedly to all types of viscometers, although the general tendencies may be the same in all instruments. When a non-polar suspending liquid is used, the electrostatic charge which can develop on the suspended particles may affect the measurement of viscosity. The nature of the electric fields produced in the suspension should be investigated for any viscometer before viscosity measurements are attempted.

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(Part 2—"The viscosity and sedimentation of suspensions of rough powders"—will appear in the December issue.)

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# A photographic method for displacement/time recording

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Small spherical reflectors attached to or formed upon the surface of a moving body produce a point image of a fixed light source. This image moves with the body to which the reflector is attached, and by recording its motion on a continuous-feed or rotating drum camera, a displacement/time record is obtained in the form of a fine and sharply focused continuous line which is suitable for detailed analysis. Recording may be carried out under normal lighting conditions by this technique. The camera described gives time scales in the range 0 to 315 in/sec.

An investigation of the kinematics of a mechanism under various operating conditions is greatly assisted by records of the displacement/time characteristics of the individual members. The method of recording must not impose any appreciable loading on the mechanism, and this is a serious restriction when the normal operating forces are small. An obvious method is cinematography at a suitable frame speed, but while this provides an excellent visual record of the performance, detailed analysis requires the plotting of curves from measurements made on the individual frames of the film. This is extremely laborious, and the accuracy is limited by the definition of the separate exposures. In the case of high accelerations, an impracticable frame speed would be necessary to obtain a reasonably accurate diagram.

In the present method, a point image of a fixed light source is formed on the surface of the moving body, and the motion of this image is recorded photographically as a continuous line. It is therefore necessary that the part of the mechanism in question, or a rigid attachment to it, should be visible for at least that portion of its cycle that is under analysis. There is no mechanical loading of the mechanism for recording purposes.

## OPTICAL SYSTEM

If a point source of light can be formed at a fixed point on the surface of a moving body, its motion can be recorded, to a base of time, on a continuous-feed or rotating-drum camera. Eyles<sup>(1)</sup> refers to the early use of flashlamp bulbs attached to the moving object for motion study, and in more recent years, this method has been used for displacement/time recording on mechanisms such as aeroplane under-carriages. As light sources for attachment to moving objects, flashlamp bulbs suffer the disadvantages of fragility, the need for wiring, and lack of definition due to the finite dimensions of the filament. The present method employs a small spherical reflector to form a point source when illuminated by light from a photoflood bulb or other intense light source. The intensity of the point source gives satisfactory photographic recording under any conditions of normal lighting other than bright sunlight, and the definition permits subsequent enlargement up to about 10 times, with a corresponding

gain in the overall accuracy. The reflectors can be dimensioned to form the point source at the surface of the object to which they are attached, if this is considered to be necessary.

The following types of reflectors have been used with success in this form of recording:—

(a) 4 BA steel bolts with  $\frac{3}{16}$  in square heads, the surface of the head being ground and polished to form a convex spherical reflecting surface of about  $\frac{3}{8}$  in diameter. (b) Bolts similar to the above, but with a concave spherical surface. These can be made by polishing a flat on the end of a steel bar, and pressing a polished steel ball into it to produce an indent. The bar can then be machined to form the complete bolt. (c) In some cases a spherical indent can be formed by pressing a steel ball on to the surface of the mechanism link whose motion is required, but the surface of the link must first be highly polished. (d) Polished ball bearings can be mounted on the moving object, or threaded on to a string (e.g. a pulley-cord) to record their motion. (e) In certain objects, such as a helical spring of round section, the surface is already formed by two radii in perpendicular planes, and although some loss of definition occurs, satisfactory records of the coil motion can be obtained from a spot of white enamel applied direct to the surface. An oil film on a black steel surface can also be used as a reflector.

## RECORDING TECHNIQUE

The time scale is introduced by recording the motion of the reflector on a continuous-feed or rotating-drum camera, and film speeds ranging up to some 25 ft/sec have been found adequate for all applications to date.

Film or paper can be used as the recording medium, the former when photographic enlargement is to follow, and the latter for projection on to a screen. In either case, the final image can frequently be displayed to give the full scale magnitude of the displacement, so that direct measurements can be made from it without the use of a scale factor.<sup>(2)</sup> A lens aperture of  $f4.5$  will cover most requirements, but it has usually been possible to use considerably smaller apertures and so eliminate background fogging when recording in natural lighting conditions.

In the case of a movement of fixed amplitude, it is useful to take a "still" photograph with the film at rest,

and so obtain a record of the line of motion. This also acts as a reference ordinate for records of movement in a curved path. A second "still" is frequently taken with a scale placed in the plane of movement to give a scale factor for subsequent enlargement. A number of reflectors can be fitted to obtain simultaneous records of the movement of different links in a mechanism, and one reflector on a stationary member provides a datum line on the record. If the reflectors can be mounted to move in paths lying along the one straight line, any ordinate drawn across the record will give the true relative positions of the reflectors at that instant of time. As most recordings can be made with apertures in the range  $f22$  to  $f11$ , the resulting depth of focus allows some latitude in the distance of the plane of motion of the individual reflectors from the camera lens, but it has hitherto been possible to limit variations in these distances so that the one scale factor applies to all traces.

In the case of a drum-camera, several records can be taken on the one length of film, if either the lens or the entire camera is traversed normal to the film to give a relative displacement of the records. The time scale can be determined from the film speed, if this is known to sufficient accuracy, otherwise timing dots can be recorded from a discharge tube or spark gap mounted in the camera.

#### DRUM CAMERA

Any form of drum or continuous feed camera designed for recording low-speed phenomena would be suitable for the technique described above. The drum camera used in the present instance consisted of a drum 10 in. in diameter driven through a two-speed reduction gearbox by a  $\frac{1}{25}$  h.p. 24 V d.c. shunt motor fitted with a potentiometer speed control,<sup>(3)</sup> giving film speeds in the ranges 0-63 and 0-315 in/sec. The lens, shutter, and focusing controls were provided by mounting a plate camera over a transverse slot in the drum housing. Timing dots at 0.02 sec intervals are obtained from a small spark gap supplied from a spark coil whose primary circuit is switched by an electrically maintained tuning-fork. This timing device is located diametrically opposite to the camera lens, and can be traversed to record at any selected point on the width of the film.

When recording periodic phenomena, it is desirable that the records of the event and the timing should not overlap, and that they should start and finish on either side of the break in the film, leaving a gap for the "still" photographs referred to earlier. These functions are carried out automatically by the cam-operated switches and solenoid arrangement shown in Fig. 1. *A* is an initiating switch mounted on the driving shaft, and remaining closed for one revolution only once it has been initially closed by operation of an external cable release. *B* and *C* are two micro-gap switches operated from cams on the driving shaft, cam *C* being shaped to give momentary opening of switch *C* during each

revolution. With *A* closed, the timing-spark circuit is complete. After a further shaft rotation of  $180^\circ$ , *B* closes and therefore the camera shutter opens and *D* closes, this lag of  $180^\circ$  being necessary because the event and timing sources are diametrically opposite. After one complete revolution switch *A* opens and remains open, thus interrupting the timing circuit, and at  $1\frac{1}{2}$  revolutions switch *C* is opened momentarily to close the camera shutter and open *D*. The cam profiles and their relative angular positions enable any desired length and position of the records to be pre-determined, and the entire recording is carried out automatically by the initial operation of the cable release bringing switch *A* into operation.

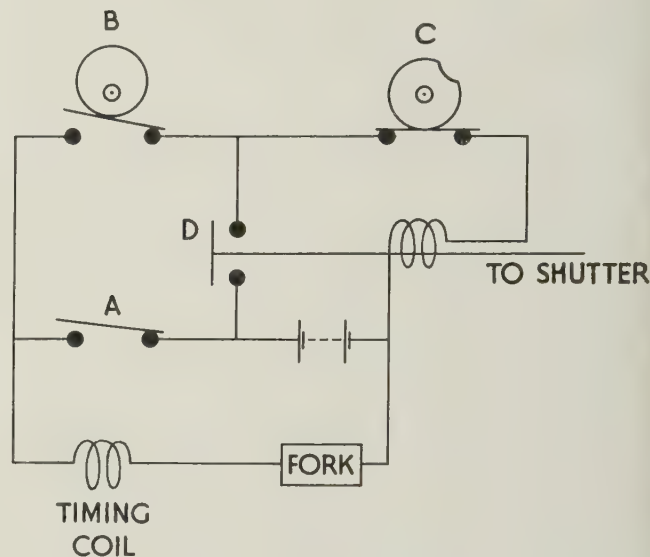


Fig. 1. Camera control circuits.

The complete camera is mounted on a stand adjustable for height, and can be rotated to set the drum axis parallel to the line of motion of the reflector bolt. The camera lens and timing spark can be traversed between recordings, so that several records can be taken on the one length of film.

#### TYPICAL RECORDINGS

Specimen records are reproduced in Figs. 2, 3, and 4. Fig. 2 records the motion of an oscillating arm carrying a reflector bolt, taken at a film speed of some 200 in/sec. The "still" photographs of the arc of motion and of a centimetre scale are shown, with timing dots towards the upper margin. This is typical of the normal recording technique.

Fig. 3 records the motion of the individual coils of a helical spring when the tension is suddenly reduced, and was obtained at a film speed of about 30 in/sec. Spots of white enamel were used to form a reflector on each coil, their initial state, with the spring in tension, being given by the horizontal lines at the right-hand



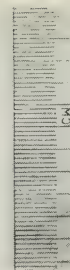


Fig. 2. Record using reflector bolts

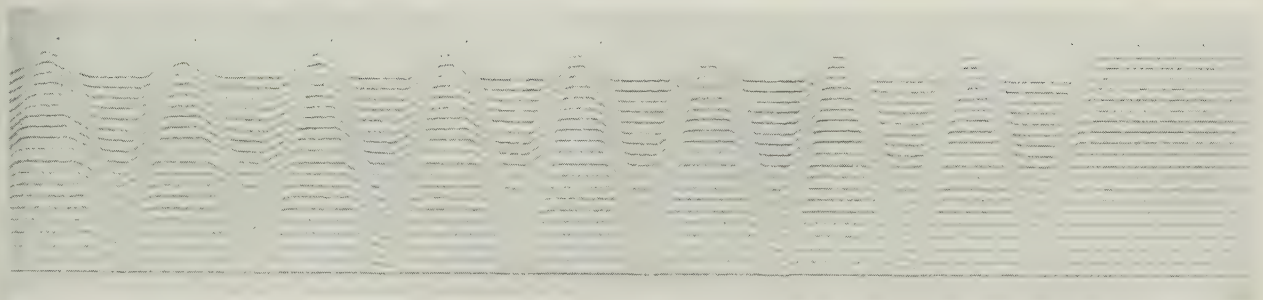


Fig. 3. Reflexions from coils of helical spring

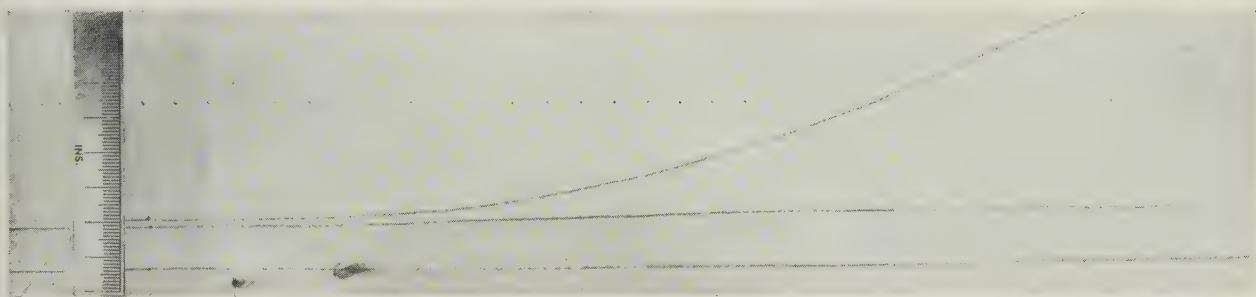


Fig. 4. Record using steel ball-bearings threaded on to pulley cords

edge of the paper. The timing dots are close to the upper margin of the photograph.

Fig. 4 is reproduced from a research programme in which the advantages of continuous-line recording as compared with cinematography have been used to extend the use of the mechanical analogue to a transmission line<sup>(4)</sup> under fault conditions or load changes. The model consists of co-axial radial arms connected by light springs at their outer extremities, and loaded with weights suspended from cords passing round pulleys that are co-axial with the individual arms and rigidly attached to them. Changes in mechanical loading correspond to variations in the electrical conditions of the corresponding network, and on a sudden change of loading, the model passes from the initial to the final state through a transient oscillation. The movements of the radial arms can be recorded by reflector bolts attached to them, resulting in a record with curved ordinates. In certain cases, as represented in Fig. 4, it is more convenient to record by reflexion from small steel ball-bearings threaded on to the pulley cords, as

this gives linear ordinates. The black smudges on the record result from the rupture of short fuse-wires that are connected to the model in such a way that the blowing of the fuse initiates the required change in loading. The instant of rupture is indicated by an intense spot in each smudge.

#### ACKNOWLEDGMENTS

The author is indebted to Mr. J. Brown and the Staff of the Electrical Engineering Department Workshop, who designed and constructed the camera and accessories described in this paper, and to Mr. G. I. Dawson, who developed the recording technique resulting in records such as that reproduced in Fig. 4.

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# The rheological properties of dielectric polymers\*

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The rheological properties of a number of dielectrics have been studied by the application of shear stress to a cylindrical tube of the material. Creep under constant stress has been measured over periods ranging from a few milliseconds to 18 months after applying the stress and the dynamic modulus and internal friction coefficient have been measured over a frequency range of  $10^{-4}$  to  $10^3$  c/s.

The results obtained indicate that the rheological properties can be expressed in terms of a distribution of mechanical relaxation times. This distribution is hyperbolic for a range of times from  $10^{-3}$  to  $10^7$  sec and enables creep and dynamic measurements to be correlated.

The increasing use of solid polymers in the combined role of electrical insulants and constructional materials has focused attention on their rheological properties, which for practical applications are perhaps almost equal in importance to their electrical properties.

The usual object is to establish relationships between stress, strain, temperature and time and to interpret them in terms of the internal structure, molecular configuration and bond strengths. The strain-time relation under constant stress is important where materials are subjected to clamping stresses, as in transformers, where creep of the dielectric can loosen clamps and cause breakdown of the transformer. The dynamic stress/strain relation is important with impulsive stresses as in transformers during electrical surges. Polythene finds extensive use in cable insulation, where its rheological properties are of great importance. Polystyrene is widely used in high-frequency circuits and in low-loss capacitors.

The object of the work described in this paper is to determine strain/time relations for a number of materials under constant stress and also under alternating stress. In the latter case results are expressible in terms of a dynamic shear modulus and internal friction coefficient. The variation of these parameters with frequency can be predicted by assuming a distribution of relaxation times, the form of distribution function being given by the strain/time relations.

Under constant stress, the strain may be divided into three components<sup>(1)</sup> as follows:—

(a) Hookean strain, which is attained immediately the stress is applied and is elastic or reversible; the strain disappears as soon as the stress is removed. The strain obeys Hooke's law (is linear with stress), hence its name, and is attributable to bond stretching and bending, and probably the stretching of weak Van der Waals "bonds" between chains.

(b) A component whose rate of increase decreases steadily with time. This is also elastic but requires time for complete recovery. It is known as primary creep,<sup>(2)</sup> elastic after-effect, high elastic deformation,<sup>(1)</sup> or recoverable strain, and is generally attributed to chain uncoiling.

(c) An irreversible component which may or may not increase linearly with time of application of stress. This is known as secondary creep or non-recoverable strain and is attributable to inter-chain slipping.

It is well known that in general any stress system is resolvable into two fundamental components, uniform tension or its negative counterpart, compression and shear,<sup>(3)</sup> but the strain which is produced by a combination of these stresses has been found experimentally<sup>(4)</sup> to be incapable of being analysed into separate components corresponding to the components of the stress. This would introduce great difficulties were it not for the fact that experience shows that the time dependence is associated largely, if not entirely, with shear strain. Pure shear strain can be provided by torsional stress for strains up to about 0.07, so that tests in torsion give results which are both amenable to mathematical analysis and capable of practical application.

## APPARATUS

For the foregoing reasons, experiments were made in torsion. Test specimens were prepared by machining the material into tubular form to the dimensions shown in Fig. 1. The material was not affected by machining

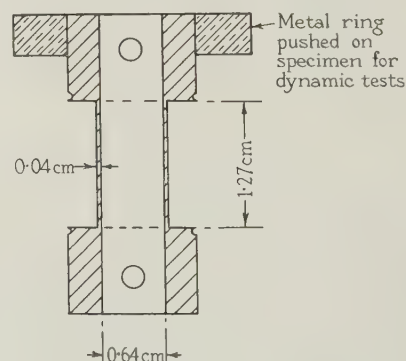


Fig. 1. Section of specimen used for tests on dielectric polymers

except in the case of polythene, which was subsequently annealed in boiling water for two hours. This restored the material to its original condition. The angle of twist is measured by reflexion of light from two plane

\* Based on reports Refs. L/T171 and L/T250 of The British Electrical and Allied Industries Research Association.



mirrors on the inner edges of the end pieces, clipped into the grooves shown in the figure.

### STATIC TESTS

The apparatus for long time tests has been described in an earlier paper.<sup>(5)</sup> With this apparatus strain can be measured in a few seconds after applying the load, and strain readings were taken at intervals for times up to 18 months after loading. After removal of the load the recovery of strain with time was observed. If the lower portion of the specimen is unclamped, recovery can take place without any friction due to the apparatus. The strain can be measured to within  $\pm 1.5 \times 10^{-5}$ .

Tests indicated that considerable creep takes place in the few seconds between loading the specimen and observing the strain. An apparatus was therefore designed by which the strain can be measured a few milliseconds after applying the stress.<sup>(6)</sup>

### DYNAMIC TESTS

The strain/time records obtained from the apparatus described in <sup>(6)</sup> show an initial oscillation which decays in a few milliseconds. The dynamic shear modulus and internal friction coefficient can be determined from the frequency and logarithmic decrement of the oscillation, provided the moment of inertia of the system is concentrated at one point. As described in <sup>(6)</sup> this was achieved by fitting a ring tightly to the top of the specimen (Fig. 1). A range of frequency was obtained by using rings of different moments of inertia and the dynamic modulus  $G$  and internal friction coefficient  $\eta$

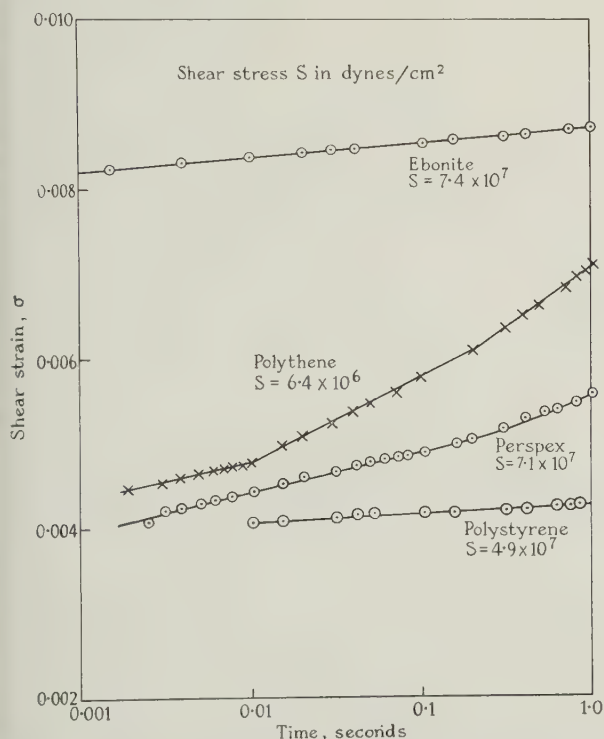


Fig. 2. Strain against log time relations for times up to 1 sec after loading

measured for frequencies from 30 to 1 000 c/s. Further tests made with a modified form of the apparatus enable the range to be extended down to  $10^{-4}$  c/s. This apparatus is also described in <sup>(6)</sup>.

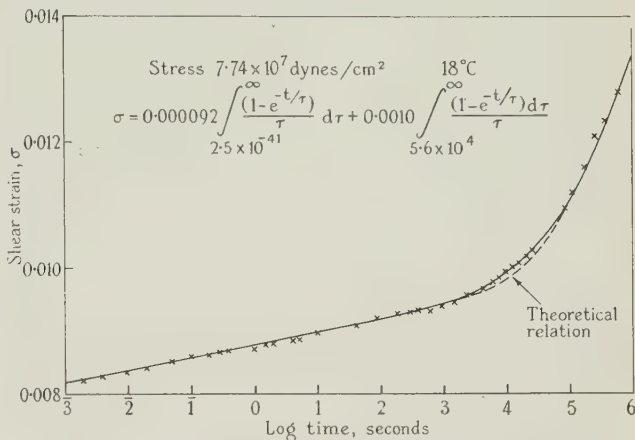


Fig. 3. Strain against log time relation for Ebonite

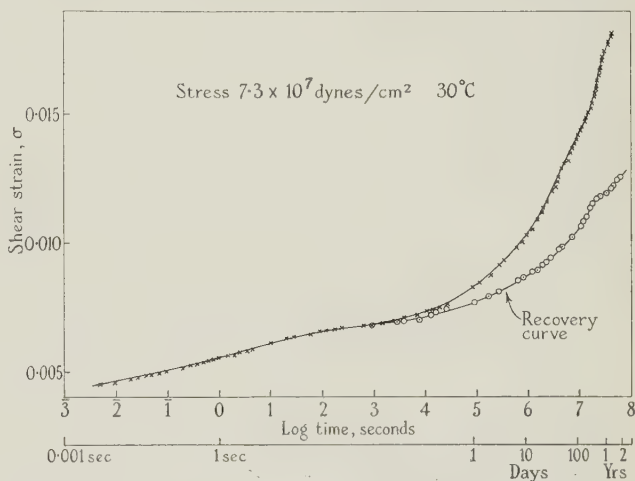


Fig. 4. Strain against log time relation for Perspex.

### EXPERIMENTAL RESULTS

**Static Tests.** Strain/log time curves obtained at 16 to 18°C for times up to one second after application of stress are given in Fig. 2. These show that considerable creep occurs during the first 1/10 sec after the application of stress. This increase of strain in the case of polythene is 32 % of the strain 1 msec after applying the stress and 23 % in the case of Perspex. The corresponding increase for ebonite and polystyrene is 3.5 %. Curves obtained at different stresses show that the strain at any given time is proportional to the stress, up to the maximum stress used,  $7.5 \times 10^7$  dynes/cm<sup>2</sup>. Figs. 3 and 4 show strain/log time relations for long times. These long time tests were made with the earlier form of torsion apparatus using weights.

Fig. 4 shows the strain/log time curve for Perspex which is similar in shape to that obtained for ebonite in Fig. 3. Polystyrene samples fracture after the load

has been applied for a few days, making long time tests impracticable. This is probably due to crazing, i.e. small cracks form in the material which considerably reduce its strength. Polystyrene is particularly susceptible to crazing. The stress was applied for 18 months in the case of Perspex. Although the strain/log time relation is similar to that for ebonite, the curve is not so smooth. An interesting feature is the absence of any indication that the strain approaches a terminating value. Creep appears to continue indefinitely as long as the stress is applied.

The recovery curve for Perspex is also plotted in Fig. 4, and shows the strain recovered after removal of stress. This curve follows the extension-time curve for several hours and then recovery proceeds at a slower rate than the corresponding extension. Two years after removal of stress the recovery has not reached a terminating value.

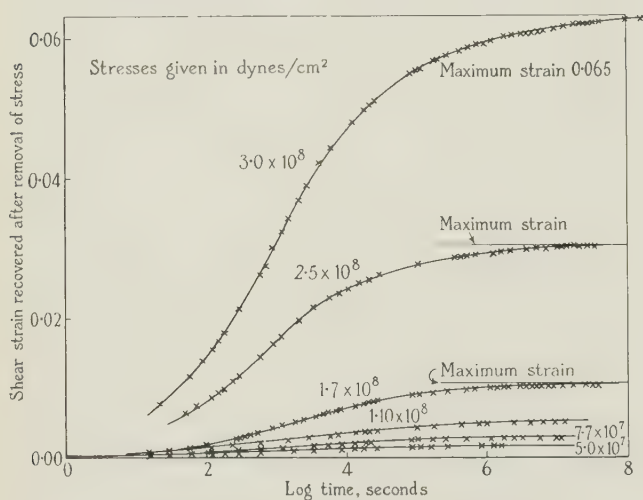


Fig. 5. Recovery curves for Perspex after loading for 1 day

A number of tests was made on samples of Perspex to which were applied for a given time loads different for each sample. The recovery curves are given in Fig. 5 (the recovered strain is here taken as the strain recovered from the value 10 sec after unloading—which is the same as the strain 10 sec after loading). The ordinates marked maximum strain correspond to the strain just before removal of stress and it is seen that non-recoverable strain is absent except at the highest stress. The strains have been separated into their three components as described in the introduction and are plotted in Fig. 6, "instantaneous strain" being taken as the value 10 sec after loading. Experiments show that primary creep occurs for times down to  $10^{-3}$  sec after application of stress; it is therefore difficult to separate it from Hookean strain so that the value 10 sec after loading was taken, this being the shortest time in which the strain could be conveniently measured with this apparatus which was that used for long time tests. The instantaneous strain is linear with stress, but recoverable strain is only linear up to stresses of about  $8 \times 10^7$

Table 1. Values of shear modulus  $G$ , and coefficient of friction  $\eta$  for dielectrics at different frequencies

Material	Log frequency, c/s	$G \times 10^9$ dyne/cm <sup>2</sup>	Log $\eta$ poise	$\tan \delta \times 10^3$
Polythene ("Akathene," grade 20 L.T.R.)	2.440	1.52	4.86	8.2
	1.960	1.39	5.43	11.1
	0.000	1.07	7.30	11.4
	1.398	0.97	7.87	12.2
	2.700	0.86	8.59	14.2
	3.256	0.64	9.98	18.0
	4.663	0.48	10.60	24.0
	4.041	0.40	11.08	20.7
Perspex (Unplasticized, polymethyl methacrylate)	2.904	19.2	5.15	3.7
	2.442	18.5	5.58	3.6
	1.654	17.2	6.53	5.6
	0.000	13.3	8.18	7.0
	2.845	11.6	9.18	5.6
	3.518	11.1	10.30	3.8
	4.380	10.4	11.40	3.6
	4.176	10.3	11.62	3.8
Ebonite (rubber 68 parts, sulphur 32 parts by weight. Vulcanized for 18 hours at 130° C)	2.788	10.1	4.90	3.1
	2.334	9.9	5.08	1.6
	1.544	9.3	5.99	2.3
	0.000	8.9	7.25	1.3
	1.518	8.8	7.74	1.3
	2.825	8.5	8.38	1.2
	3.342	8.4	9.78	1.0
	4.518	8.2	10.82	1.7
	4.000	8.1	11.40	2.0
Polystyrene ("Distrene" con- taining 2.4% of methanol soluble material)	2.848	12.9	4.25	0.6
	2.373	13.0	4.88	0.8
	1.601	12.3	5.54	0.7
	0.000	12.1	7.40	1.3
	1.518	12.0	7.80	1.1
	2.826	11.7	8.40	0.9
	2.601	11.7	8.58	0.8
	3.380	11.6	9.79	0.8
	4.416	11.2	10.75	0.8
	4.080	11.2	10.85	0.5

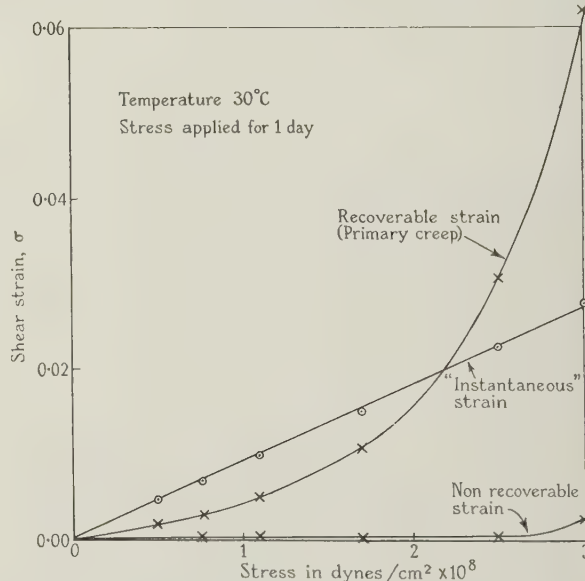


Fig. 6. Analysis of stress against strain relations for Perspex

dynes/cm<sup>2</sup>, and then increases very rapidly with stress above this value. Non-recoverable strain appears only at the highest stress which corresponds to 315 kg/cm<sup>2</sup>, and is then only 2% of the total strain. The shear strength of this material (Perspex) was measured and found to be approximately 500 kg/cm<sup>2</sup>, so that non-



recoverable strain was only just observed when the stress was about 60% of the breaking stress.

Similar results were obtained for ebonite and polythene.<sup>(4)</sup> The strain/log time curve for polythene is near, but the line always shows abrupt changes in slope (Fig. 7).

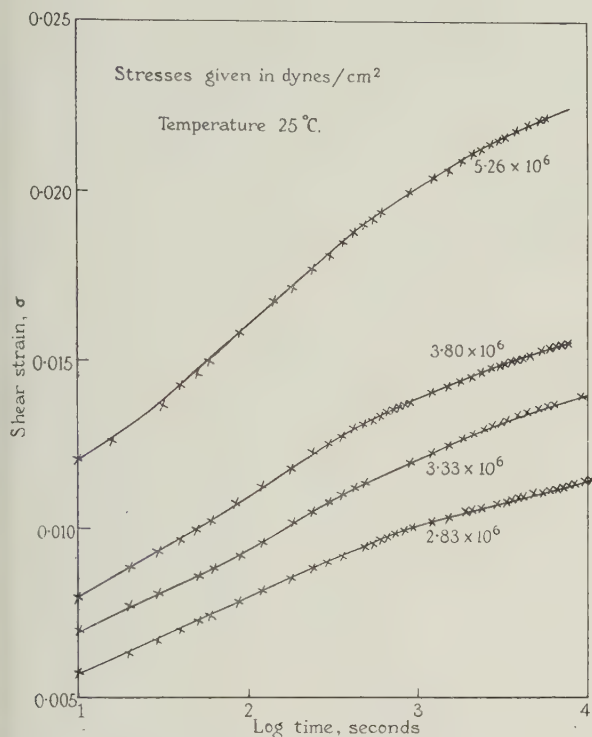


Fig. 7. Strain against log time relations for Polythene

**Dynamic tests.** Experimental results are given in Table 1. Tests at frequencies up to 1 c/s give the loss angle  $\delta$  directly; from which  $\eta$  is calculated, using the relation  $\tan \delta = \eta\omega/G$ . At frequencies greater than 10 c/s,  $\eta$  is obtained directly from the decay of oscillation as described earlier, whence  $\tan \delta$  can be calculated. The table shows that the dynamic modulus  $G$  increases with frequency as would be expected since less creep occurs at high frequencies. The coefficient of internal friction  $\eta$ , however, decreases rapidly with frequency.

#### DISCUSSION OF RESULTS

Experiments have shown that for small stresses, i.e. values up to  $8 \times 10^7$  dynes/cm<sup>2</sup> for Perspex (Fig. 6), the stress strain relations both for instantaneous and recoverable strains are linear. As will be shown later, experimental results can therefore be expressed in terms of a distribution of relaxation times. The nature of the distribution function will be calculated from the strain/time relation at constant stress. In 1943 some authors believed this to be a very difficult, if not impossible task to do generally (see Ref. (2), p. 71), but already in 1944 Whitehead<sup>(7, 8)</sup> has given an exact procedure, and later Gross<sup>(9)</sup> summarized various methods, together with

additional contributions. Nolle,<sup>(10)</sup> who finds a wide frequency spectrum for rubber, has used an approximate method whose validity has not been established. All these methods, if applied to the experimental curves given in this paper, will give the distribution function that has been obtained, but the treatment given here has the advantage that the functions employed are much simpler than those required by other methods.

The strain/log time curves of Figs. 3 and 4 are made up of two linear portions. If the mechanical behaviour can be represented by a Kelvin or Voigt element, i.e. a spring and dashpot in parallel, the strain at time  $t$  after applying the stress can be expressed in the usual form.<sup>(11)</sup>

$$\sigma = (S/G)(1 - e^{-t/\tau}) \quad (1)$$

where  $G$  is the modulus of the spring and  $\eta$  the viscosity of the dashpot,  $\tau$  being the relaxation time given by  $\eta/G$ . Relaxation time is here defined as the time for the strain to fall to  $1/e$  of its value when the stress is removed. This applies to a single Kelvin element. Equation (1) does not, however, represent the curves of Figs. 3 and 4. Assuming that the behaviour corresponds to a very large number of Kelvin elements in series, of which the number having relaxation times between  $\tau$  and  $\tau + d\tau$  is  $f(\tau) d\tau$

$$\text{then} \quad \sigma = S \int_0^{\infty} f(\tau)(1 - e^{-t/\tau})d\tau$$

where  $f(\tau)$  is termed the distribution function of relaxation times.

The slope of the  $\sigma/\log t$  curve is given by

$$\frac{\partial \sigma}{\partial \ln^* t} = S \int_0^{\infty} \frac{t}{\tau} f(\tau) e^{-t/\tau} d\tau$$

In particular, if  $f(\tau) = k/\tau$ , where  $k$  is a constant

$$\frac{\partial \sigma}{\partial \ln^* t} = Sk \int_0^{\infty} \frac{t}{\tau^2} e^{-t/\tau} d\tau = Sk$$

If  $f(\tau) = k/\tau$  the slope is then constant, i.e.  $\sigma$  and  $\log t$  are linearly related. Although the foregoing integral is finite  $\sigma$  becomes infinite if  $f(\tau) = k/\tau$  holds at  $t = 0$ . In fitting the constant  $k$  to the observed results the distribution is assumed to cut off at some very small value  $\tau_0$ . Provided the shortest observed time is much longer than  $\tau_0$ , the preceding relations are unaffected, since, if  $t \gg \tau_0$ , changing the limit from 0 to  $\tau_0$  has a negligible effect on the integral  $(t/\tau^2)e^{-t/\tau}d\tau$ .

Figs. 3 and 4 suggest that the strain can be expressed in terms of two distributions. The curve for ebonite (Fig. 3) is found to fit the equation

$$\sigma = 9.2 \times 10^{-5} \int_{2.5 \times 10^{-41}}^{\infty} \frac{1 - e^{-t/\tau}}{\tau} d\tau + 10^{-3} \int_{5.6 \times 10^4}^{\infty} \frac{1 - e^{-t/\tau}}{\tau} d\tau \quad (4)$$

except over the curved portion joining the two straight lines.

\*  $\ln$  is the Napierian logarithm.

The relationship for polythene shown in Fig. 7 cannot be expressed exactly by a series of the form  $\int_{\tau_1}^{\tau_2} \frac{k(1 - e^{-t/\tau})}{\tau} d\tau$  which gives a series of straight lines joined by curves and not sharp discontinuities.

## DYNAMIC TESTS

Static tests show that the strain/time relation for plastics is expressible in the form

$$\sigma = S \int_{\tau_0}^{\infty} \frac{k(1 - e^{-t/\tau})}{\tau} d\tau = \phi(t) \quad (5)$$

Where  $\tau_0$  is very small compared with the shortest time of measurement.

(We are now considering small values of  $t$  so that the second term of equation (4) can be omitted.)

Using the well-known Hopkinson superposition principle

$$\sigma = \int_{-\infty}^t \phi(t-u) \frac{dS(u)}{du} du \quad (6)$$

and applying an alternating stress at zero time

$$S(u) = S_0 \sin \omega u \quad \text{when } u > 0 \\ = 0 \quad \text{when } u < 0$$

$$\sigma = S_0 \int_0^t \int_{\tau_0}^{\infty} \frac{k}{\tau} [1 - e^{-(t-u)/\tau}] \omega \cos \omega u d\tau du$$

on substituting for  $\phi(t)$  from (5).

Evaluating the integral with respect to  $u$ ,

$$\sigma = S_0 \int_{\tau_0}^{\infty} \left( \frac{\sin \omega t}{1 + \omega^2 \tau^2} - \frac{\omega \tau \cos \omega t}{1 + \omega^2 \tau^2} + \frac{\omega \tau e^{-t/\tau}}{1 + \omega^2 \tau^2} \right) \frac{k d\tau}{\tau}$$

If  $t$  is large enough, the transient term is negligible and

$$\sigma = S_0 k \int_{\tau_0}^{\infty} \frac{d\tau}{\tau(1 + \omega^2 \tau^2)} \sin \omega t - S_0 k \int_{\tau_0}^{\infty} \frac{\omega d\tau}{1 + \omega^2 \tau^2} \cos \omega t \quad (7)$$

The strain in a Kelvin element whose modulus and viscosity are given by  $G$  and  $\eta$  respectively for a stress  $S = S_0 \sin \omega t$  can readily be seen, as in the electrical analogue, to be given by

$$\sigma = S_0 G (G^2 + \omega^2 \eta^2)^{-1} \sin \omega t - S_0 \omega \eta (G^2 + \omega^2 \eta^2)^{-1} \cos \omega t \\ = (S_0/G) \sin \omega t - (S_0 \tan \delta / \omega \eta) \cos \omega t$$

if the loss angle  $\tan \delta = \omega \eta / G \ll 1$ .

Comparison with equation (7) yields

$$\frac{1}{G} = k \int_{\tau_0}^{\infty} \frac{d\tau}{\tau(1 + \omega^2 \tau^2)} \\ = k \ln [\omega \tau / (1 + \omega^2 \tau^2)^{1/2}] \Big|_{\tau_0}^{\infty} \\ \simeq -k \ln \omega \tau_0 \quad (8)$$

if  $\omega^2 \tau_0^2 \ll 1$

and

$$\frac{\tan^2 \delta}{\eta \omega} = k \int_{\tau_0}^{\infty} \frac{\omega d\tau}{1 + \omega^2 \tau^2} \\ = k\pi/2 \quad \text{if } \omega \tau_0 \ll 1 \\ \eta = k\pi G^2 / 2\omega \quad (9)$$

i.e.

Differentiating equation (8)

$$d(1/G)/d(\ln \omega) = -k$$

The reciprocal of the shear modulus should therefore decrease linearly with the logarithm of the frequency.

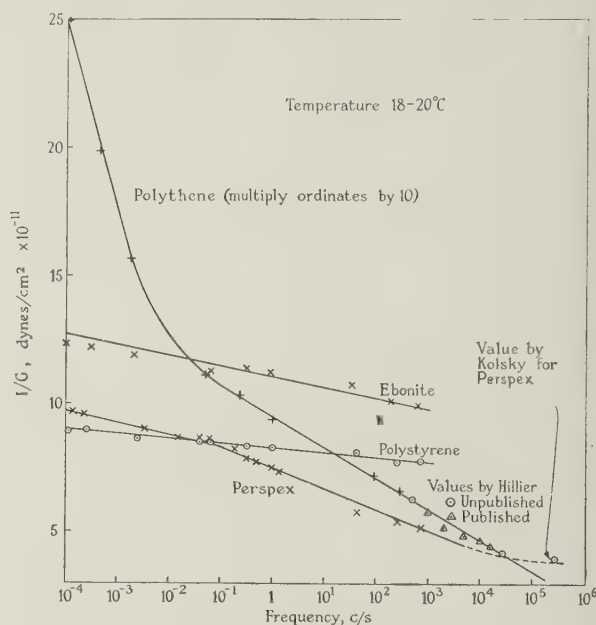


Fig. 8. Relation between reciprocal of dynamic shear modulus and logarithm of frequency

Fig. 8 shows experimental results where the linear relation is obeyed for ebonite and polystyrene. With Perspex the line changes slope at  $10^{-1}$  c/s. This seems connected with an upper limit  $\tau_1$  in the distribution function which is indicated by the reduction in the slope of the  $\sigma/\log t$  curve of Fig. 4 in the region of  $\log t = 2.2$ . Assuming the existence of an upper limit, equation (8) becomes

$$1/G = k \ln \left\{ (\tau_1/\tau_0) [(1 + \omega^2 \tau_0^2)/(1 + \omega^2 \tau_1^2)]^{1/2} \right\}$$

which reduces to equation (8) when

$$\omega^2 \tau_1^2 > 1 > \omega^2 \tau_0^2$$

If  $\tau_1 = 5$  sec,  $\omega^2 \tau_1^2$  decreases rapidly with frequency as the latter is reduced from 0.1 to 0.025 c/s ( $\log f$  reduced from 1.0 to 2.4), and if  $f < 0.02$  c/s,  $1/G$  becomes independent of  $f$  and the  $1/G/\log f$  curve becomes horizontal. In Fig. 8 the slope is reduced considerably below  $10^{-1}$  c/s. The distribution does not end sharply at a specified value because the  $\sigma/\log t$  curve of Fig. 4 does not become horizontal around  $\log t = 2$ , so that the slope of the  $1/G/\log f$  curve of Fig. 8 will not become zero for frequencies below 0.1 c/s. The value given by Kolsky<sup>(12)</sup> at high frequencies is obtained from



measurements of the velocity of the head of a pressure pulse in the material and it is estimated that this corresponds to a frequency of about  $3 \times 10^5$  c/s.

The  $1/G/\log f$  curve for polythene is linear only at the upper and lower frequencies. A linear relation over the whole range could not be expected in view of the broken nature of the strain/log time curves of Figs. 2 and 7. The slope of the  $1/G/\log f$  curve is greatest at the lower frequencies as would be expected in view of the increased slope of the  $\sigma/\log t$  curve for time above 0.01 sec (Fig. 2). The values given at frequencies above 500 c/s were obtained by Hillier<sup>(13)\*</sup> who measured the velocity of longitudinal waves along polythene threads. The shear modulus is deduced from Young's modulus by multiplying the latter by the usual factor  $1/[2(1 + \mu)]$ , where  $\mu$  = Poissons' ratio. Equation (9) shows that  $\eta\omega$  is independent of frequency if  $G$  is constant. Fig. 8 and Table 1 shows that  $G$  increases slightly with frequency so that  $\eta\omega$  should also increase.  $\log \eta$  is plotted against  $\log$  frequency in Figs. 9 and 10, which show a linear relation. The slope of the line is slightly greater than  $-1$ , which means that  $\eta\omega$  increases slightly with frequency. The increase of  $G$  with frequency is slight for polystyrene and ebonite, so that the slope of the line should be nearly  $-1$ , which is the case. The loss angle is given by

$$\tan \delta = \eta\omega/G \\ = k\pi G/2$$

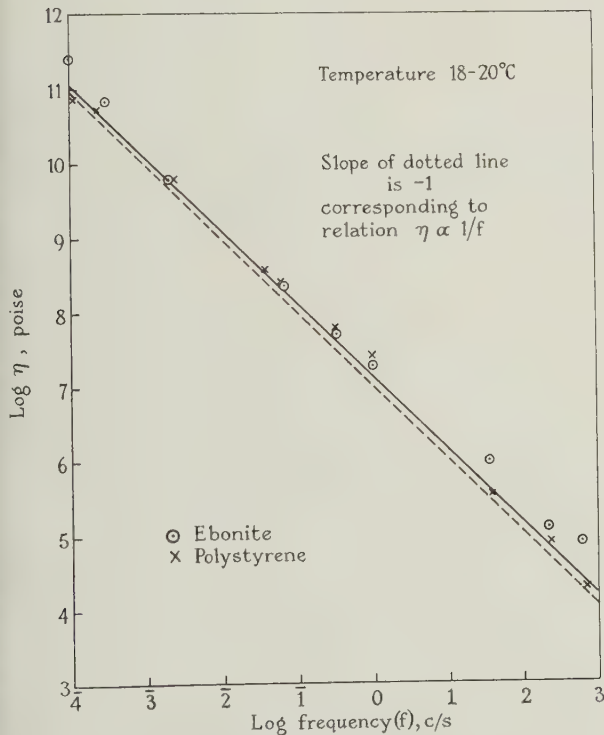


Fig. 9. Relation between  $\log \eta$  and  $\log$  frequency for Ebonite and Polystyrene

\* The author is indebted to Dr. Hillier for permission to quote from the unpublished results.

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from equation (9) and should increase slightly with frequency because  $G$  also increases. The values of  $\tan \delta$  given in Table 1 are rather irregular, but they remain substantially constant over the whole frequency range covering seven decades.

#### CORRELATION BETWEEN STATIC AND DYNAMIC TESTS

Equation (9) gives the dynamic viscosity  $\eta$  in terms of the slope of the  $\sigma/\log t$  curve at constant stress. Table 2 gives values of  $\eta$  at 1 c/s which were calculated from equation (9), and it is seen that these values agree fairly well with the experimental values.

Table 2. Comparison of dynamic values of  $\eta$  with those calculated from static data

Material	$b \times 10^{12}$ (Fig. 4)	$G \times 10^9$ (Table 1) dynes/cm <sup>2</sup>	Value of $\eta \times 10^7$ at 1 c/s Calculated poise	Observed (Table 1)
Polythene..	220	1.1	2.9	2.0
Perspex ..	9.8	13.3	18.7	15.0
Ebonite ..	2.25	9.0	2.0	1.8
Polystyrene ..	2.5 (Fig. 2)	12.0	3.9	2.5

\*  $b$  is the slope of the  $\sigma/\log t$  curve and is  $2.3k$  where  $k$  is the slope of the  $\sigma/\ln t$  curve.

#### EVALUATION OF THE EXPLICIT STRAIN/LOG TIME RELATION FROM THE DISTRIBUTION FUNCTION

The strain time relation is expressed as

$$\sigma = S \int_0^\infty f(\tau)(1 - e^{-t/\tau})d\tau$$

Expressing  $f(\tau)$  as  $k/\tau$  gives

$$\frac{\sigma}{Sk} = \int_{\tau_0}^\infty \frac{1 - e^{-t/\tau}}{\tau} d\tau \\ = \ln T_0 - \int_{\epsilon \rightarrow 0} \left( \ln \epsilon + \int_{\epsilon}^\infty \frac{e^{-T} dT}{T} - \int_{T_0}^\infty \frac{e^{-T} dT}{T} \right)$$

putting  $T = t/\tau$ .

The last term is negligible since  $T_0 > 10^6$ . Now if  $\epsilon$  is small,

$$-Ei(-\epsilon) = \int_{\epsilon}^\infty \frac{e^{-T} dT}{T} = -\gamma - \ln \epsilon$$

where

$$\gamma = 0.577 \text{ (see reference }^{(14)}),$$

so that

$$\sigma/Sk = \ln T_0 + \gamma \\ = 2.3 \log t/\tau_0 + 0.577$$

This gives a linear strain/log time relation whose slope is given by  $2.3k$  and whose intercept at  $t = 1$  is given by

$$Sk[2.3 \log (1/\tau_0) + 0.577].$$

#### INTERPRETATION OF FORMULAE IN TERMS OF MOLECULAR MECHANISMS

Equation (4) expresses the strain/time relation for ebonite in terms of two distributions. The lower limit of the distribution in the first integral,  $= 2.5 \times 10^{-41}$  sec,

is much shorter than could be expected for any real relaxing element, which could not move in less than some  $10^{-13}$  sec, the period of a molecular oscillation. The reason for the discrepancy cannot be determined without tests at times shorter than  $10^{-13}$  sec, but it is evident that either the distribution  $1/\tau$  fails at short times, the population of relaxing elements rising much faster than  $1/\tau$  as  $\tau \rightarrow 0$ , or else there exists a third group of elements with relaxation times not widely distributed, and all very much shorter than  $10^{-3}$  sec. The response occasioned by bond stretching and bending as distinct from chain uncoiling, may well behave thus. If  $10^{-3}$  sec is somewhat arbitrarily taken as the true lower limit for the distribution which determines the left-hand end of Fig. 3, then its total contribution to the strain at  $10^{-3}$  sec is about 0.002, leaving a strain of about 0.006 to be contributed by elements with a

relation between molecular structure and macroscopic behaviour. If the suggestions made in the last paragraph are justified, the mechanisms of strain in ebonite (at room temperature and low stress) is as follows:—

Some two-thirds of the strain observed at 1 sec is due to bond stretching and bending which is completed certainly within  $10^{-5}$  sec. Over a range of times extending in both directions beyond  $10^{-3}$  and  $10^4$  sec there is a distribution varying as  $1/\tau$  which contributes about one-third of the observed strain. This response presumably comes from chain uncoiling. Above  $10^4$  sec a third type of response begins to make an appreciable contribution to the strain. This, so far as it has been measured, seems also to be distributed as  $1/\tau$ . It may be interpreted as corresponding either to breakage of primary bonds (possibly the  $-S-$  bond in ebonite) or to release of interchain entanglements. It would make a very large contribution to the strain if measurements were continued long enough. Its contribution in the short interval  $10^5$ – $10^6$  sec is already equal to the whole contribution from chain uncoiling.

The difference between polythene and the other materials tested may be significant in view of the fact that polythene is a crystalline polymer with some degree of rubber-like properties; while the others are amorphous polymers. The strain/log time relation for rubber under heavy load has been observed to consist of a series of broken lines.<sup>(15)</sup>

#### CONCLUSIONS

It is shown that creep occurs in dielectrics under constant stress, and although the rate of creep diminishes with time it does not become zero, which means that the amount of creep does not reach a terminating value. Thus many dielectric structures are never completely stable. The creep is wholly elastic and may be represented by a system of Kelvin elements in series, each element having a relaxation time  $\tau$ . The distribution function of relaxation times is expressible in the form  $k/\tau$  for values of  $\tau$  from  $10^{-3}$  to  $10^5$  sec, the range of times over which tests have been made. For Perspex this distribution has been found to be approximately true for values of  $\tau$  up to  $10^7$  sec. Each relaxation time corresponds to the uncoiling of a chain and is related to chain length and other factors.

The reciprocal of the dynamic modulus decreases linearly with the logarithm of the frequency and the internal friction decreases inversely as the frequency: both these results are shown to be consequences of a distribution function having the form already deduced from the static tests.

#### ACKNOWLEDGMENTS

The author is indebted to the Director of the British Electrical and Allied Industries Research Association for permission to publish this paper, and to Mr. R. D. Keeler for assistance in making some of the measurements.

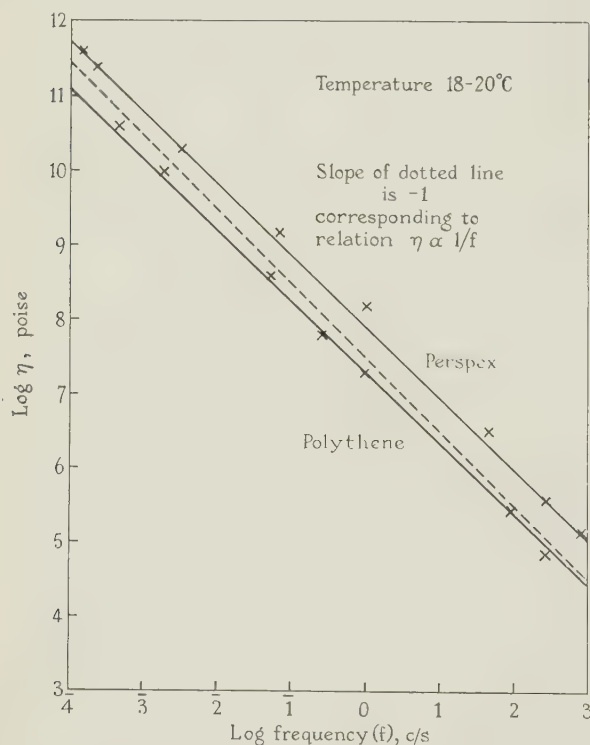


Fig. 10. Relation between  $\log \eta$  and  $\log$  frequency for Perspex and Polythene

restricted range of very short relaxation times. This response is certainly complete before  $10^{-3}$  sec, or it would appear as a curvature at the left-hand end of Fig. 3, which is in fact quite straight. It must therefore be somewhere in the range  $10^{-13}$  to  $10^{-5}$  sec.

The nature of the distribution function at these very short times is of no importance as regards the practical behaviour of materials at times longer than  $10^{-3}$  sec, since the response of the short time elements is complete at longer times and has no further effect on the slope of the strain/time curves. The question is, however, of theoretical importance as a means of interpreting the



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## Correspondence

## The Pirani Effect in a Thermionic Filament as a Means of Measuring Low Pressures

I have been unable to reconcile the results given in the paper entitled "The Pirani Effect in a Thermionic Filament as a Means of Measuring Low Pressures," which appeared in your May issue, with other experiences in this subject, and I should like to offer the following comments.

In the figure are given calculated and measured values of  $\Delta V/V$ , the fractional change of voltage required to maintain constant filament temperature when the pressure in a vacuum system changes from zero to the amounts marked on the curves.  $\Delta V$  is proportional to the energy removed by molecules impinging on the filament, i.e., to the Pirani effect. Calculations were based on the classical equation for heat loss from a hot wire.<sup>(1)</sup> The experimental values were determined by standard bridge technique. Values given by Dunoyer<sup>(2)</sup> are even lower than those obtained in this laboratory. It will be observed that corresponding values of  $\Delta V/V$  deduced from the May paper are, in general, about one hundred times higher than those in the curves. Furthermore, their values for hydrogen are about 5 times smaller than for air, whereas the more usual value is about 1.6 times greater.<sup>(3)</sup>

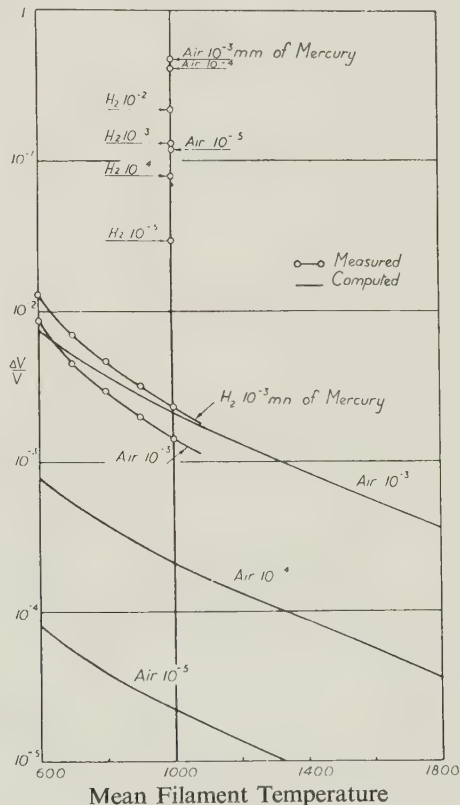
A more practical criticism of this paper arises if the filament temperatures quoted are compared with the values given in Langmuir's tables.<sup>(4)</sup> According to Langmuir, the thermionic emission from pure tungsten at 1000° K is  $10^{-15}$  A/cm<sup>2</sup>. This is about  $10^{12}$  times smaller than observed by the authors. Again, if a temperature of 2400° K was needed for 20 mA emission, the same tables show that at 90  $\mu$ A emission the temperature would be 2000° K. The stated filament operating temperature, 1000° K, would thus appear to have been underestimated, though a possible alternative explanation of the discrepancy is that the filament was of thoriated rather than pure tungsten. The observed variations of operating voltage could then be explained as consequences of variation of filament work function with gas composition, pressure, and heat treatment. Variation of work function could also account for the wide divergence of the "zero" voltage required for the initial points of the air and hydrogen calibrations and for the fact that the shapes of the curves in the paper bear no relation to the shapes predicted by the heat loss equation.

It seems to me most probable that, by neglecting the

dominant role played by work function in studies of the emission from tungsten, the authors have been led to draw erroneous conclusions as to the effect responsible for their observations.

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Trafford Park, Manchester.



Fractional change of voltage required to maintain constant temperature when pressures change from "zero" to the values marked on the curves. Values calculated from results published in May issue are marked on the 1000° K ordinate.

As I alone was responsible for the paper to which Mr. Blears refers I am replying alone to his criticism. I am grateful to him for raising two important points. First, the change in surface character of the filament with pressure will clearly affect the results, e.g., via work function changes, and it should have been made clear that the extension of the work mentioned in the paper is in fact an attempt to investigate the influence of adsorbed gases. Secondly, Mr. Blears has pointed out that the emission from tungsten would not account for the observed results. Since, however, the temperature was less than  $1000^{\circ}\text{K}$ , the conclusion is that the filament was not pure tungsten. This would not alter appreciably the Pirani effect to be expected. I have calculated two points on the  $1000^{\circ}\text{K}$  ordinate after Roberts<sup>(5)</sup> using emissivity  $\epsilon_t = 0.1$ <sup>(6)</sup>, and accommodation coefficients for air and hydrogen 1.0 and 0.25 respectively. I find values which are (a) for air at  $10^{-3}\text{ mm}$  four times the computed value shown by Mr. Blears and (b) for hydrogen at  $10^{-3}\text{ mm}$  four times the experimental value shown by him. Agreement between observation and calculation will largely depend on the assumed values of these three constants.

Difficulty arises in assessing the observed values of  $\Delta V/V$

determined by a bridge method without details of the experimental procedure. The importance of securing a standard filament surface condition before each heat loss observation has been stressed in the past, and in a recent paper Bremner<sup>(7)</sup> shows how very large errors may arise due to thermal effects present in methods frequently used.

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## NOTES AND NEWS

### New Books

**History of Physics.** By MAX VON LAUE. (Academic Press, Inc.) 1950. Pp. viii + 150. Price 17s. 6d.

There is room and need for a short history of physics, and Max von Laue's volume admirably fills this gap.

There are many ways of writing a history of physics but two only of them need be considered here. One way may be illustrated by the *History of Physics* due to the late Florian Cajori in which the division of physics into its classical groups and the advances made in all the groups are traced century by century. It happens that significant discoveries have been made about the turn of each century if we give a generous interpretation to the phrase "turn of the century." But the method is to be deprecated.

Another way consists in recognizing that the history of physics is bound up with advances in chemistry, astronomy and engineering and to take certain selected topics and to trace the developments in each topic through the centuries. This method, which has been successfully employed in Dr. Buckley's *Short History of Physics*, is the one employed by von Laue, who divides his history into sections which are devoted to the following topics: Measurement of Time, Mechanics, Gravitation and Action at a Distance, Optics, Electricity and Magnetism, the Reference System of Physics, the Bases of the Theory of Heat, the Law of Conservation of Energy, Thermodynamics, Atomistics, Nuclear Physics, Physics of Crystals, Heat Radiation and Quantum Physics.

These topics are treated with a deceptive simplicity of language and it is only on scrutinizing this treatment with great care that one realizes that the treatment is not likely to be fully appreciated by that difficult fellow the intelligent layman, and the reader should possess a certain maturity of mind and a knowledge of physics of what we may call Vith-form standard fully to appreciate the good *provand* which von Laue has provided.

It is not to be expected that, within the limits of 150 pages, the author can discuss minor topics in any detail, but he has managed to include a surprising number of accounts of travels along the by-paths of scientific advances which add interest to the story of the advances in physical science.

It can be said of Max von Laue that he has taken a large share in these recent advances, and there are few historians better qualified than he, who has borne a large share in the labour and conflict of the day, to describe and assess the magnitude and direction of those advances.

It may seem ungrateful to point out what are in the reviewer's opinion some trifling slips in the impressive story which he has told so well. Thus, in an account of the discovery of the planet Neptune, he gives the whole credit of the discovery to Leverrier and the name of Adams is not even mentioned. In discussing the invention of logarithms he puts the claim of Joost Burgi on a level with that of Napier. This hardly seems justifiable in view of the fact that Napier's work was so much more important, complete and constructive. He points out, also, that there is some justification for speaking of what we in England call Boyle's Law as Boyle-Mariotte's law, and justifies Mariotte's claim to originality though Boyle had antedated him by no less than fourteen years, by remarking that there was very little intercommunication between scientific men of this period. That may very well be, but the curious reader will find in the appendix to Tait's *Properties of Matter* (third edition, 1894) a discussion of a passage in the *Principia* in which Newton writes, in a highly sarcastic manner, of Mariotte's labours in an allied problem. Again in considering the discovery by Stevinus of Bruges of the law of the inclined plane, he merely states the fact and gives no account of the manner in which Stevinus arrives at his discovery.

In discussing the manner of Fahrenheit's methods of taking the zero of his scale of temperature as the temperature



f a mixture of ice and salt he has overlooked the recently published correspondence between Fahrenheit and Boerhaave\* in which it would seem that Fahrenheit determined his zero point by taking the interval between the melting point of ice and the normal human body temperature, and prolonging the scale to a distance below the ice-point equal to half this difference.

The author's short account of Davy's experiment on friction and the nature of heat is misleading. Davy's experiments were concerned with the friction of two pieces of ice which were rubbed together *in vacuo*. Similar experiments were carried out with wax, but von Laue describes the experiments in the sentence "Davy . . . , by means of a clock-work, . . . caused two pieces of metal to rub against each other in a vacuum." Practically all writers on heat since Davy's day have been content to describe the results as authoritative and place them on a level with the famous experiment of Rumford on the boring of cannon. It was left to Andrade† to point out the serious deficiencies in Davy's experiment.

These are, however, but minor matters and it is to be hoped that, in the new edition which will most certainly be called for in the near future, the author will add to the value of the book by including therein some account of the applications of natural philosophy that have resulted in the foundation of such bodies as the National Physical Laboratory in England and the corresponding one at Charlottenburg in Germany.

ALLAN FERGUSON.

**Light and Sound.** By M. NELKON, M.Sc., A.K.C. (London: William Heinemann Ltd.) Pp. vii + 342. Price 11s. 6d. net.

Perhaps the first consideration by which a teacher assesses a textbook on elementary optics is the sign convention chosen by the author. Since any convention is ultimately only an economy of mental effort, there seems much to be said for using the very simplest where this will suffice for the work in hand, and changing to a Cartesian convention where circumstances require it—much as a clarinettist changes instruments when fingering or transposition become difficult. This is the line taken by Sir Cyril Ashford in a recent book, and is really that adopted by Mr. Nelkon, who has used the real-is-positive convention throughout the text, and given a full account of the "new Cartesian" convention in an appendix. This book covers the geometrical optics required for the advanced standard in the new certificate examination in a concise and satisfactory manner. Three chapters on Sound then follow, an arrangement which shows that the author has considered another important teaching point—how much of the general theory of wave motion should precede the study of physical optics. Newton's study of sound was probably responsible for turning the attention of Huygens to the crude beginnings of a wave theory of light that had been made by Hooke; so it seems appropriate as well as desirable to approach the wave theory with a fair understanding of sound, and this sequence is commendable. But one feels that it might have been more fully exploited, for the general wave-motion work has been subordinated to a factual treatment directed to syllabus requirements in sound. The first of the two chapters on physical optics which conclude the book is too brief to satisfy those who prefer at this stage

to recapitulate much of the work of geometrical optics using a wave treatment, but again this will suffice for syllabus needs.

The obvious errors in the book are all slips of general scholarship rather than of physics. It is surprising to read that Romer's work was ignored by the scientists of his time, for its effects were immediate and widespread; that Foucault measured the velocity of light in water; that Michelson died in 1936; and that Newton proposed a corpuscular theory of light "about 1660," a date which would certainly render consistent the statement that this was "prior to the wave theory of light," since it is some years before the publication of the *Micrographia*. This is no trivial fault-finding, for though the errors may individually be small they add up to an impression that the author has worked within fairly wide tolerance limits for these matters. This is a pity, for if physics is to be esteemed as an integral part of a liberal education, its historical aspects must surely be presented in a way that the historian can respect.

G. R. NOAKES

**A Problem in Chemical Engineering Design: The Manufacture of Mononitrotoluene.** Edited by J. M. COULSON and F. E. WARNER. (London: The Institution of Chemical Engineers.) Pp. 55. Price 15s. 6d.

This book is based on four lectures demonstrating how a chemical engineering design problem should be tackled, the example chosen being the manufacture of *o*- and *p*-mononitrotoluenes, involving a large number of unit operations, such as liquid-liquid contacting, the transfer of heat, distillation, crystallization and centrifuging. The business of the chemical engineer when planning his work in the initial stages is to make sure that he has overlooked nothing which is relevant to his problem, and as an illustration of the very wide range of subjects which he must consider, and of the number of angles from which each single item of equipment must be viewed, this monograph is excellent. The formulae and methods of calculation chosen for the various steps in the design could in one or two instances have been more up-to-date, but those adopted are quite adequate for the purpose of the book.

Although this book is primarily of interest to the chemical engineer, the general reader could derive benefit from reading it. It will give him a good picture of the chemical engineer's workshop, and if he is a physicist or physical chemist, he will be fascinated by the large number of points of contact between his subject and chemical engineering.

P. DOCKSEY

**Advances in Electronics.** Vol. II. Edited by L. MARTON. (New York: Academic Press, Inc.) Pp. x + 378. Price 59s. 6d. net.

This second volume of *Advances in Electronics*, which commenced with Volume I in 1949, amply justifies continuation of the series and gives every promise that a yearbook of this nature will be of real value.

The extent to which international co-operation has been fostered in the volume under review is shown by the fact that of the nine authors, three are American, four British, one Canadian and one French. The editor and editorial board are to be commended on this, and it is to be hoped that future volumes will continue in the same spirit; by spreading the authorship in this way there is no doubt that balanced reviews are presented to the reader. A brief

\* *Annals of Science*, 2, p. 133 (1937).

† *Nature*, p. 359, 9 March, 1935.

summary of the ground covered by the several articles is all that space will allow.

H. Moss, in a most useful review, traces the progress made in cathode-ray tubes over the past decade and notes differences in design adopted in British, American and Continental practice, data not readily obtainable elsewhere; his sections dealing with design methods will be welcomed by all tube designers.

A treatise on electron lenses follows by P. Grivet, who deals with this important subject under the main divisions of electrostatic lenses and magnetic lenses, their defects and methods of correction, and finally gives examples of some typical lenses used particularly in electron microscopes and cathode-ray tubes.

The companion paper by G. Liebmann on field plotting and ray tracing in electron optics reviews the several numerical methods that are or have been used. The aim in this work is to study the fields produced by electrodes of a given geometry and to determine the particle trajectories through them or, alternatively, to evolve suitable shapes of electrodes or pole pieces to give a desired trajectory.

G. F. J. Garlick's survey of cathodoluminescence is also of importance in cathode-ray tube work. The paper summarizes the types of practical cathode-ray tube screens currently in use, experimental studies of their electrical characteristics and stability under electron bombardment, the theoretical interpretation of cathodoluminescence phenomena and a discussion of the present trends in research in this subject.

Behaviour of dielectric materials under electric stress is of wide interest and importance, and H. Frölich and J. H. Simpson have chosen the subject of intrinsic dielectric breakdown in solids for discussion. The theory is carefully built up and, as the authors point out, agreement with experimental results indicates that it accounts for the essential features of such phenomena.

In "The Microwave Magnetron," G. Hok sketches the state of to-day, the mechanism of oscillation, properties of the magnetron oscillator, compared with other microwave oscillators, and gives a short account of recent contributions to the further development of the magnetron towards meeting more exacting demands in performance.

Two other papers follow dealing with microwaves. The first, by G. T. Rado, deals with ferromagnetic phenomena at microwave frequencies. A survey is made of dispersion of permeability, resonance absorption and hysteresis effects and the account of the first of these is extended to semiconductors. As the author states, many gaps exist in our present knowledge and such a survey as this is more than useful in revealing these clearly. The second paper is by D. K. Coles and reviews microwave spectroscopy. This is an entirely new branch of spectroscopy, made possible by developments in microwave techniques, and is concerned with the absorption in materials, mainly gases, at sharply defined frequencies between about 10 000 and 100 000 Mc (wavelengths 3 cm to 3 mm).

A. J. MADDOCK

## Journal of Scientific Instruments

### Contents of the November issue

#### ORIGINAL CONTRIBUTIONS

- A compensating circuit for the Simmons shielded hot-wire anemometer. By L. L. Fox, P. L. Palmer and D. Whittaker.
- A device for maintaining a constant potential for electro-chemical work. By F. W. Chambers.
- A tunable audio-frequency amplifier of variable selectivity. By E. A. G. Shaw.
- An experimental synchronous contactor for repeated automatic operation. By H. W. Baxter.
- A circuit for the automatic stabilization of X-ray tube current. By C. S. Lees and M. D. Armitage.
- An equipment for the microscopic examination of metals and crystals in polarized light at temperatures from  $-130$  to  $+35^{\circ}\text{C}$ . By J. C. Monier and R. J. Hocart.
- Apparatus for the study of creep of dielectric polymers and their dynamic rheological properties. By W. Lethersich.
- An instrument to demonstrate the synthesis of a wave-guide mode of propagation. By H. M. Barlow.
- A simple manometer for use in measuring low air velocities. By D. B. Spalding.

#### LABORATORY NOTES

- A cover-slip carrying case. By D. G. Drummond.
- A constant-level device for liquid air. By G. F. H. Box and M. S. Walker.
- Voltage variation of lead-acid accumulators at small discharge rates. By F. A. Benson and D. Harrison.
- A vernier method of using a slide-rule. By M. J. Everitt.
- A bridge method for the control of electrolytic polishing. By L. F. Bates and C. D. Mee.

#### NOTES AND NEWS

#### CORRESPONDENCE

The electrical viscometer—R. McKennell and A. R. Boyle.

#### NEW BOOKS

- Radio communication at ultra-high frequency—Metallurgical applications of the electron microscope—The theory and design of inductance coils—Sketching for craftsmen—Dictionary of engineering and machine shop terms—Glossary of terms used in electrical engineering.

#### NOTES AND COMMENTS

## British Journal of Applied Physics

Special Reports and Original Contributions accepted for publication in future issues of this Journal

SPECIAL REPORT ON INSTITUTE OF PHYSICS' CONFERENCES  
High-intensity X-ray Beams—London 1950.

#### ORIGINAL CONTRIBUTIONS

- Errors in diffusion measurements by the Loschmidt method. By L. Tordai.
- An apparatus for studying the magnetic field due to an electric current in a long straight conductor. By H. Owen.
- Factors limiting the accuracy of electrolytic plotting tanks. By P. A. Einstein.
- Replica techniques in electron microscopy. By A. W. Agar and R. S. M. Revell.
- A new method of preparing paste and powder specimens for the electron microscope. By F. Schofield.
- The application of factor analysis to tests on cheese. By R. Harper and M. Baron.

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## SPECIAL REPORT

## Summarized proceedings of conference on high-intensity X-ray beams—London, April 1950

The Annual Spring Conference of the X-ray Analysis Group of The Institute of Physics was held in London on 13-14 April, 1950. The subject was "The production and application of high-intensity X-ray beams." In addition to the evening discourse delivered by Prof. A. Guinier of the Faculté des Sciences et Conservatoire National des Arts et Métiers, Paris, sixteen papers, together with a number of shorter contributions, were given during the four sessions; an exhibition of apparatus and photographs was also held. The discourse, and other contributions, and the discussions are summarized in this report.

## HIGH-INTENSITY X-RAY GENERATORS

*Moving Targets*

The first session was opened with an historical review by DR. A. TAYLOR (Mond Nickel Co. Ltd.) on "The development of the high-powered moving-anode X-ray tube." By moving the anode relative to the electron stream to increase the rate of heat dissipation, the continuous beam intensity of an X-ray tube can be greatly enhanced, either by increasing output for a given focus, or by reducing focal area. In early models by Vood and Crookes,<sup>(1)</sup> the tube itself rotated about a fixed cathode, or the electron beam was magnetically deflected. Modern practice prefers the moving anode. In the sealed-off models of Thomson and Coolidge,<sup>(2)</sup> early prototypes of modern medical therapy tubes,<sup>(3)</sup> the anode was rotated by means of an induction motor with external stator, Coolidge also introducing hot-wire high-vacuum cathode electron emission. Vacuum-tight flexible bellows have also been used to give oscillatory<sup>(4),(5)</sup> or gyratory<sup>(6)</sup> motion; but greater power is achieved in demountable tubes with high-speed rotating targets, classical models with characteristic moving vacuum seals being the Müller-Clay machines of the Royal Institution<sup>(7)</sup> (stuffing-box seal), the Leeds mercury-seal rotors,<sup>(8)</sup> the French (greased-cone) rotor,<sup>(9)</sup> and the Russian models<sup>(10)</sup> with the rotor of a Siegbahn-Gaede molecular pump as target.

Most of the above are limited to a single radiation end, at most, two windows. The speaker concluded with a description of two models, designed and built by himself, possessing advantages in multiple windows and radiations, and extreme simplicity, ease and speed in control and servicing. One, an open-bowl model,<sup>(11)</sup> cooled by low-vapour-pressure oil acting as heat exchanger, dispenses even with a moving seal; the other<sup>(12)</sup> (Fig. 1), designed to fit a commercially produced unit, incorporates four windows and the "Gaco"<sup>(13)</sup> seal, with an alternative multi-metal sectored target. It was stressed that several designs exist only in patent specification, and that while theory approximates to practice with many metals (iron, copper, etc.), the useful manganese, if plated, strips by thermal stress on normal foci at maximum tube currents as low as 10 mA.

The leviathan of X-ray generators is the 50-kW Müller-Clay rotor<sup>(7)</sup> of the Davy Faraday Laboratory. Designed for continuous operation at full power, its

22-cm anode now normally rotates at 1 300–1 500 r.p.m., the hexaphase rectified circuit supplying a smooth d.c. input from 2 A at 25 kV to 1 A at 50 kV.

DR. D. P. RILEY (Royal Institution, London), in the next paper, "The modified 50-kW X-ray generator at the Davy Faraday Laboratory," indicated recent developments made to achieve maximum efficiency in useful X-ray production. Target surface is true to 0.0025 mm, a new h.t. rectifying system has been installed, and an automatic voltage regulator and current stabilizing device are being incorporated. Focusing was discussed in detail. Application of the approximate Müller formula<sup>(14)</sup> (which neglects lateral cooling and re-entrant path) to a radial rectangular line focus of length  $a$  and breadth  $b$  on a target revolving at 1 300 r.p.m. leads at full safe power of 50 kW to the relation  $a\sqrt{b} = 0.55 \text{ cm}^{3/2}$ . Maximum safe power input  $W \propto a\sqrt{b}$ ; brilliancy  $P$  ( $\equiv W/\text{focal area}$ )  $\propto 1/\sqrt{b}$ . Clearly, for a given  $W$ , a highly asymmetric focus of low  $b$  tends to high  $P$ .

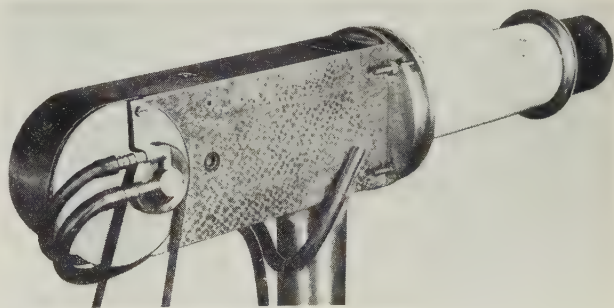


Fig. 1. Medium-power high-intensity rotating-anode X-ray generator

Is the brilliancy utilized? A too extended pseudo-focus introduces the "umbrella" effect or is cut off by slits; a more equi-dimensional one involves too great a foreshortening below the optimum angle of  $2.5^\circ$  off grazing emergence, while, with  $a$  approaching 60 mm, terminal focal speeds are very different. Electron ray tracing analysis using the rubber sheet method<sup>(15)</sup> has led to a new cathode system giving even emission free of "hot spots," end effects, and the depth effects of helical or thick wire filaments. The new filament of tungsten ribbon  $1.0 \times 0.1 \text{ mm}$  in section is maintained taut exactly 6 mm behind the front surface of an

unbiased focusing slot  $6 \times 25$  mm and is claimed to yield  $a$  25,  $b$  0.1 mm with negligible tail and no wings, giving  $P = 9$  kW/mm<sup>2</sup> at  $W = 23$  kW. Lateral cooling permits some exceeding of the calculated Müller limit. The generator now operates  $\sim 7$  h per day, usually at  $1/2$ – $2/3$  maximum power and normally, but not invariably, at its finest focus of  $b = 0.1$  mm.

DR. W. H. TAYLOR (Cavendish Laboratory, Cambridge) briefly indicated the connexion between the four Cambridge papers, which had their inception in the need for a fine clean powerful beam for direct examination of polycrystalline metal. Mr. Gay had extended the study of monochromators and surface aberrations following the early collaboration of Dr. R. C. Evans (Dept. of Mineralogy, Cambridge) and the late Mr. J. N. Kellar (Cavendish Laboratory); Mr. Hirsch had made the slit system his main study; while work on the generator itself, launched by Mr. Kellar, had been continued by Messrs. Hirsch and Thorp, the latter of whom, in addition to independent developments, had been responsible for erection and test.

The generator, now available for general work, was described by MR. J. S. THORP (Cavendish Laboratory, Cambridge) in "A high-intensity X-ray generator." The anode (supplied by Metropolitan-Vickers Electrical Co. Ltd.) is a water-cooled bevelled disk of 10-in diameter rotated at 1 400 r.p.m., the vacuum and cooling-system Wilson seals<sup>(16)</sup> having a satisfactory life of 300 h on a smooth shaft. The electron gun, due to Mr. Kellar, comprises line or strip tungsten filament insulated from a slotted hood given, as grid, a variable negative biasing voltage by passing tube current through a variable resistance between grid and filament. Vacuum bellows allow precision adjustment of filament relative to both hood and anode. Both a.c. and d.c. potentials are applicable up to 5 kW. An exhaustive practical study<sup>(17)</sup> by means of pin-hole photographs illustrated the dependence of focal region and beam current  $i$  on such variables as type, geometry and relative setting of filament and hood; h.t. voltage  $V$ ; filament current  $I$ ; anode-grid distance  $D$ ; depth of filament in hood  $d$ ; and especially biasing voltage  $v$ . The main findings may be summarized as: For a given grid and filament, foci of similar dimensions (width  $b$ , length  $a$ ) are attainable by various combinations.  $b$  decreases with decreasing  $D$  or increasing  $d$  or  $v$ . Independent of  $V$ , and largely of grid but not of  $d$ , minimum focal width  $w = 50 \mu$  at  $v$  of 500–1 000 V beyond which  $i$  reaches saturation limited rather by space-charge than temperature.  $i$  increases with decrease in  $d$  and  $D$  and increase in  $V$ . Smoothing of intra-focal fine structure is favoured by high  $v$ .

Even at the finest foci, the gun can supply sufficient beam current to melt the target. A typical loading of 25 mA at 40 kVp on a focus  $1.5 \times 0.15$  mm shows a gain in brilliance as against normal sealed-off tubes of 50 times; the decrease in exposure time may be much less and depends on the type of photograph.

Attempts were made to explain the focal regions on

electron-optical principles using electrolytic trough<sup>(18)</sup> and ray tracing methods. For zero bias and varying  $d$ , qualitative agreement between calculated and observed fine structure is obtained assuming absence of space-charge, and zero thermal energy of emission of electrons limited to radial emission of uniform density. Quantitatively the observed  $b$  are much greater (7–2 mm as against 2–0.2 mm calculated) than is attributable to thermal energy and mutual repulsion, and probably space-charge is responsible. These three features are certainly operative under heavy bias conditions when the assumptions are in any event no longer approximately applicable.

"A survey of recent work in Holland" was then given by DR. J. S. WOLDRINGH (Royal Dutch Shell Group, Amsterdam). Salient features of the five Dutch rotating-anode crystallographic X-ray tubes described in the Table on page 307.

More exact analysis (Oosterkamp,<sup>(19)</sup> Woldringh<sup>(20)</sup>) of surface temperature distribution and cooling conditions has determined optimum anode wall thicknesses (copper 1.5, iron 2.0 mm) and has been used by Sangster<sup>(21)</sup> in calculating thermal stresses in anode surfaces. These show that plastic deformation is possible, to cause the "feathering" (minute cracks  $> \frac{1}{2}$  mm deep along the beam path) which develops after a few hundred hours and may reduce the theoretical maximum specific output by a factor of 2–3. The worn surface tools off as powder, but is replaceable by an adherent electrolytically-deposited layer. Synthetic rubber is favoured for the rotating-anode seals; these need anchoring, or rotational friction lowers the vacuum from  $10^{-6}$  to  $10^{-4}$  mm mercury. In Model 5, seal gases are trapped. For both static and moving targets, d.c. steady voltage permits the highest safe specific input—for rotating-anodes to a marked degree. Three-phase rectification is preferred to condenser-smoothed h.t. which is uneconomical, and dangerous at high voltages. During continuous running the cathode seating may be red-hot; this not only restricts the upper-limiting h.t. voltage, but causes defocusing through relative displacement of filament and hood by differential expansion. This is adjustable in models 4 and 5; improved thermal conduction in the latter restricts the maximum cathode end temperature to 400°C even when 200 W is dissipated in the filament. The cathode is replaceable as a unit; the anode is swivelled at 45° for easy disassembly. Adequate cooling of windows and seating is imperative, as some 30% of the electron energy is extra-focal.

High-voltage surge phenomena are troublesome; these arise between h.t. generator and X-ray tube and can be damped along with commutative oscillations by insertion of RC circuits in parallel with the X-ray tube. Lengthy routine experience has shown the value of careful material selection, regular servicing, and full maintenance of complete safety devices whose initial cost is far outweighed by the gain in output and man-hour obtained by full automatization and unattended night



*Features of Dutch rotating-anode X-ray tubes*

Generator No.	1	2	3	4	5
Location	Histological Laboratory, University of Amsterdam	Physics Dept., University of Groningen	Philips Research Laboratories, Eindhoven	Technical Physics Laboratory, Technical University of Delft	
Responsible	G. C. Heringa	D. Coster A. Brill H. de Lang	W. J. Oosterkamp	J. S. Woldringh	J. J. Arlman M. Sangster H. G. de Winter J. S. Woldringh
Operation	Since 1940	Still experimental	Laboratory, and experimental	1938. Since 1943 run > 10 000 h	1946. To be completed in 1950
Anode					
Diameter, cm	20	20	20	8	30
Nature	Cu	Cu, cyl. wall	Cu, cyl. wall	Cu, Fe, Cr, Ag	Cu, cyl. wall
p.m.	300	1 000–2 000	1 500	2 400	2 000
Vacuum seal	Liquid mercury	Oil film	Simmer ring + intervac.	Modified Müller-Clay	As No. 4 with vapour trap
Normal power	60 mA at 60 kVp	10–15 kW at 30 kV(mean)	300 mA at 40–45 kVp	100 mA at 45 kVp	500 mA at } 15–25 62.5 kVp } kW
Rect.	Mechanical Full-wave 1-phase	Smoothed Full-wave 1-phase	Full-wave 1-phase Medical type Air-cooled	Full-wave 1-phase + RC filter circuit	Full-wave 3-phase 6-valve smoothed. As E.N.R.A.F. medical installation
Local spot	Spot Diam. 5 mm	Line 0.5 × 20 mm <sup>2</sup>	Line 0.8 × 10 mm <sup>2</sup>	Line 0.8 × 10 mm <sup>2</sup>	Line 1.0 × 10 mm <sup>2</sup>
Windows	1	1	2	1 (Li or Al)	3. One for Monochromators
Cooling	Water	Water	Water + forced air	Water	Water
Cathode	As Leeds		Oblique	Includes two adjusting bellows	As No. 4
Vacuum				mm mercury 10 <sup>-5</sup> –10 <sup>-4</sup>	As No. 4
Pumps				Mercury diffusion	N.R.C. H <sub>2</sub> Oil Diffn. 50 l/sec
Safety devices				Fully protected	De luxe. Fully automatic. Shock-proof
Features			One window for Geiger counters	Full-wave 3-phase 6-valve h.t. installg.	For continuous night work unattended

operation. Dr. Woldringh concluded with a tribute to his co-operative team of scientists and technicians.

*Fine Foci*

Extremely high X-ray beam brilliancy (but low total power) is securable without anode motion if the source can be made so very small that radial cooling makes a major contribution to safety. Such beams have increasing fields of application and may dispense with an anterior slit.<sup>(22)</sup> This was the approach made by DR. R. WITTY and MR. P. WOOD (Metropolitan-Vickers Electrical Co. Ltd., Manchester) in "A micro-focus X-ray tube," and by DR. W. EHRENBURG and MR. W. E.

SPEAR (Birkbeck College, London) in "A fine-focus X-ray tube." The problem is essentially one of electron optics.

The former authors, recognizing from the Müller formula<sup>(14)</sup> for maximum safe specific input  $W$  that  $W \propto 1/(\text{linear dimensions})$  of a rectangular focus of fixed proportions, aimed at a brilliancy gain of 100 by reducing the conventional  $10 \times 1$  mm focus to  $0.1 \times 0.01$  mm. At 50 kV this should yield  $W$  of 32 (8.4) kW/mm<sup>2</sup> for a maximum target surface temperature of 1 080°C (300°); i.e. a tube current of  $\sim 0.5$  mA. Following Goldsztaub,<sup>(23)</sup> this is done in two equi-dimensional stages using a biased electron gun

with electrostatic focusing followed by an electromagnetic lens. With the co-operation of Messrs. M. E. Haine and J. S. Thorp, suitable components were designed and tested to permit a convenient tube lay-out (Fig. 2). The gun, fed at 50 kV by shock-proof h.t. cable, consists of a line filament of 0.2 mm diameter tungsten wire accurately set by screw adjustment in a slotted ( $5 \times 1$  mm) cup negatively biased by a few hundred volts. Following test combinations of filament current, bias voltage

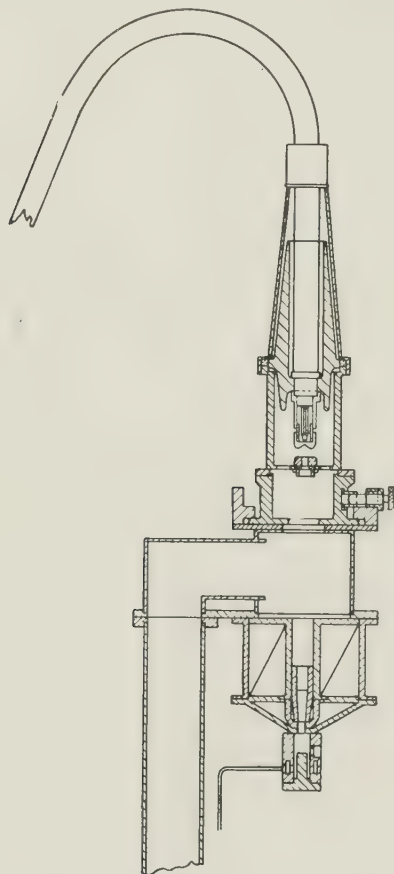


Fig. 2. Micro-focus X-ray tube—sectional view

and depth of filament in cup, to secure a high ratio of collected to emitted current (60–70%), an optimum system was found to give a focal width (Fig. 3)  $< 10 \mu$  for a precise range of lens current, maintained by a 2-valve stabilizer. Orthogonal adjustments facilitate accurate centring of the beam in the lens aperture. The target, insulated and slightly inclined to the horizontal, is kept in place by a screwed ring, only its housing needing water-cooling. Protection devices are a minimum. All vacuum joints are Neoprene rings. A valve and by-pass allow speedy change of filament and target. Although Müller's theory indicates a maximum safe beam current of 0.38 mA ( $T = 1080^\circ \text{C}$ ) at 50 kV, it is possible to run at 0.50 mA, 50 kV for 30 minutes before pitting occurs on a focus determined by microscope and pin-hole methods as  $50 \times 10 \mu$ , equivalent to at least a temporary value of  $W$  of  $50 \text{ kW/mm}^2$ .

DR. EHRENBERG, aiming at extreme simplicity, rejected conventional solutions for brilliant beams. The small focus practically nullifies the extensive engineering and high power of moving anodes; good results are obtainable by electromagnetic lens methods, but disabilities include cost and complexity of power unit for stabilized h.t. and lens current, auxiliary voltages, long filament-anode path with consequent sensitivity to stray fields and some lack of accessibility.

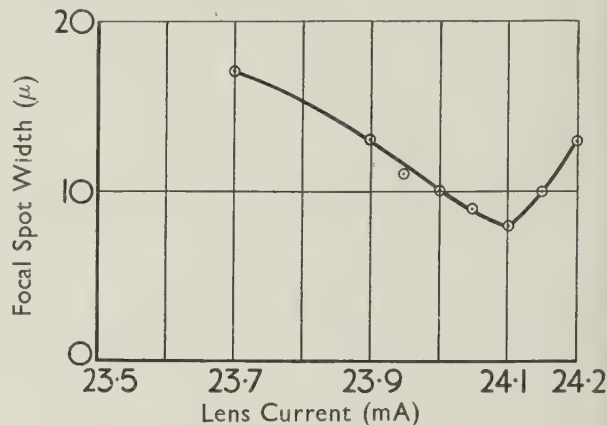


Fig. 3. Relation between focal-spot width and lens current. Bias resistance 400 kΩ, h.t. 50 kV, Target current (approx.) 0.5 mA, filament depth 14 mil.

An elegant solution of the electronics problem is arrived at simply by reversing the usual sense of the h.t. as between cathode, casing and anode in an electrostatic lens system (Fig. 4). Study of potential distributions and electron trajectories from a hooded hairpin filament taking into account off-axis distance, and velocity and angle of emission, shows that with earthed casing and cathode and h.t. anode, large cross-over and fractional magnification are securable in a short path. In practice, owing to a small group of high-velocity electrons

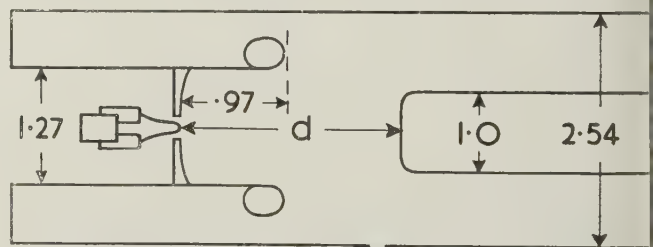


Fig. 4. Section through focusing system of fine-focus X-ray tube. Dimensions in centimetres

the anode is optimally displaced from the image plane to that of a circle of least confusion. Focal spot is markedly dependent on filament-grid setting and grid-anode distance  $d$ , but not on grid bias ( $\sim 50 \text{ V}$ ) or beam current up to 0.5 mA. Focal area radii and approximate intensity distributions (Fig. 5) determined for various values of  $d$  by pin-slit photographs<sup>(24)</sup> show that much of the energy concentrates in spots of diameter 80–20  $\mu$ . With a copper anode at 50 kV and beam



currents of 0.3–0.5 mA, average loadings of 8 kW/mm<sup>2</sup> with central peak of 11 kW/mm<sup>2</sup> and maximum surface temperatures of  $\sim 450^\circ\text{C}$  give only minor pitting over year's test.

The finished model (Fig. 6) fits directly by cone joint to its oil diffusion pump. It incorporates nickel cathode cylinder, ceramic filament-holder, bellows-adjustable filament and alternative anodes. The h.t. anode is cooled by centrifugally pumped carbon tetrachloride delivered by flexible plastic tubing. Micro-radiographs are of excellent definition. Powder photographs, especially by Seemann-Bohlin technique, take only a few minutes exposure by special micro-camera.

Both micro-focus X-ray tubes were demonstrated.

MacArthur focusing system. Tungsten strip filament is found unsatisfactory.

DR. W. A. WOOSTER (Dept. of Mineralogy, Cambridge) enquired whether the gas-discharge source, as in the Swiss electron microscope,<sup>(25)</sup> had been tried. Dr. Ehrenberg, in reply, said that a *high-power* gas-discharge type formed Müller's first model.<sup>(26)</sup>

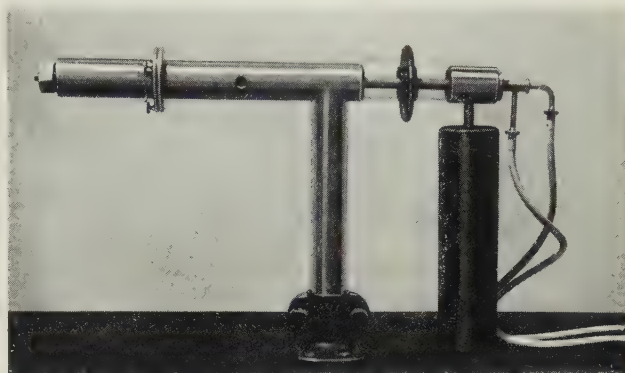


Fig. 6. Fine-focus X-ray tube—general view

PROF. SIR LAWRENCE BRAGG (Cavendish Laboratory, Cambridge) found it surprising that anti-cathode rotation is ineffectual with minute foci. Dr. Ehrenberg said it was a question of the relative rates of heat outflow and presentation of new target surface; DR. I. MACARTHUR (Dept. of Biomolecular Structure, Leeds) instanced the micro-time parallel to micro-space, where cooling, let alone rotation, is dispensed with in "flash" tubes<sup>(27)</sup> despite loadings measured in thousands of amperes.

The need for objective standards of comparison and "figures of merit"<sup>(28)</sup> was expressed by SIR LAWRENCE BRAGG, DR. WOOSTER (who recommended measurement in  $r$  units by modern Geiger-counter equipment), and by PROF. J. D. BERNAL (Birkbeck College, London), who preferred as criterion a standard substance under standard conditions; DR. A. TAYLOR doubted the full significance of factors of merit without complete layout and application. Continuing, Prof. Bernal said that the important thing is *useable* beam intensity (ergs sec<sup>-1</sup> mm<sup>-2</sup>), the user's criterion being minimum exposure time for a given performance. Small foci allow of increasing contrast effective at lower photographic densities, though their low-angle scattering performance is sensitive to "wings." Their effective use will involve the rational re-design of ancillary apparatus—a matter for the Technical Panels of the Group—and may alter the nature of the time problem to one of the nuisance of micro-working and adjustment. As in the infra-red field, wide provision of speedy instruments will attract workers. The present limiting factor is the early development of anode hair-line cracks. The fact that Prof. Wyart's experience shows these to be a function of the metal and not of "burning-out" suggests a metallurgical problem possibly to be solved by reduction of thermal strain with super-pure single-crystal targets.

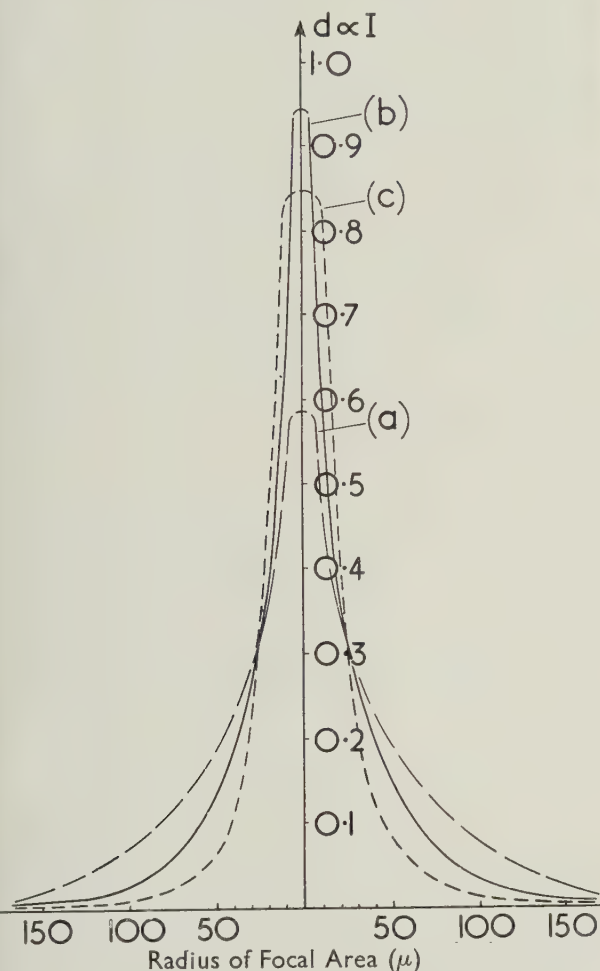


Fig. 5. Variation in width and quality of focus with grid-anode distance  $d$

(a)  $d = 2.36$  cm; (b)  $d = 2.23$  cm; (c)  $d = 2.04$  cm.

#### Discussion on Papers on High-Power X-ray Tubes

DR. R. W. H. SMALL (Chemistry Dept., Birmingham) briefly described his 12-in diameter rotor with copper target which takes up to 100 mA. Main features are Gaco seals,<sup>(13)</sup> the effective beam-path water-cooling system adapted from Woldringh, and the Astbury-

Outlining the relevance of his thermal stress work to anode cracking, DR. M. SANGSTER<sup>(21)</sup> (Royal Dutch Shell Group, Amsterdam) stated that temperature jump, not temperature gradient or maximum, is the operative factor. Under usual conditions, the thermal stresses arising in the copper anode are twice those permissible without fracture. There is no gain in using beryllium-copper and poorer conductors are still worse; but possibly better conditions prevail in micro-focus tubes.

DR. V. VAND (Lever Bros. and Unilever Ltd.) stressed the advantages of high-conductivity copper and especially of single crystals. DR. W. J. OOSTERKAMP (Philips Research Laboratories, Eindhoven) confirmed the advantages of the latter, instancing experience at Delft.

DR. A. TAYLOR preferred the intermediate size of power-tube and was perturbed by the economics of the high-power class with its drain through special mains leads, transformers and rectifiers, safety devices, maintenance, and especially anode replacement. He advocated the use of the Gaco seal, low-power drive, and adjustable clamped filaments. In reply, DR. WOLDRINGH stressed the large factor of improvement in safe brilliancy involved in rectified, especially three-phase, supplies, quite apart from surge-damping and automatization. Cathode heating was admittedly troublesome.

MR. P. B. HIRSCH (Cavendish Laboratory, Cambridge) confirmed routine difficulties in cathode assembly heating, especially with screw holds, and suggested fan cooling.

DR. D. P. RILEY attributed his trouble-free cathode, despite a 20–50 kW input, to the nature and deep setting of the filament. He preferred aluminium stearate as stuffing-box lubricant to a wearing rubber seal. His experience of diffuse scattering using power and ordinary tubes showed that brilliancy was not the sole factor. In reply to Dr. Wooster, who believed a molten target of low vapour pressure had been used to obviate anode cracking, Dr. Riley said that a routine model had apparently not been constructed.

DR. I. MACARTHUR was impressed by the present interest in power X-rays and the range of features and components now available for assembly in routine models to suit different classes of application. The stimulus of the Group's initiative in earlier meetings<sup>(29)</sup> was being justified, particularly in the advances in applied electronics. The real problem of the very small focus involved not only isolation, size, and intensity contour, but also invariant location. Its main advantage is reduced if it is subject to flicker or wander through sensitivity to field variation (even by leaks or uneven pumping) or (in agreement with Prof. Bernal) through tedium in adjusting small things. For these reasons, in initiating medium-power sharp-focus generators at Leeds, they had compromised at a moderate focal gain to ensure routine performance. He still believed the percentage gain secured by rotating targets even with

micro-foci justified their use, which in any event spread risks.

DR. J. W. JEFFERY (Birkbeck College, London) did not envisage difficulties with the Ehrenberg model whose proper field of application would be, not with fine slit but with properly designed cameras, divergent beam and Seemann-Bohlin technique.

#### THE ROLE OF HIGH-INTENSITY BEAMS IN THE PROGRESS OF X-RAY ANALYSIS

In his evening lecture PROF. A. GUINIER reviewed the production and application of high-intensity X-ray beams. Noting that, by comparison with electron diffraction, diffracted X-ray beams were of very low intensity (of the order of  $10^{-4}$  that of the incident beam) he stressed the importance of increased sensitivity for the discovery of new phenomena which are otherwise likely to remain unobserved.

Considering firstly methods of detection, it seems clear that, due to the quantum nature of radiation, increased sensitivity is unlikely. Thus an increase in the sensitivity of photographic film<sup>(30)</sup> necessitates an increase in grain size, considerable increase in exposure time is not practicable in most cases and Geiger counters are already nearly 100% efficient. Electronic means of increasing the sensitivity of detection by the use of a fluorescent screen have been used,<sup>(31)</sup> but here again no great increase can be expected.

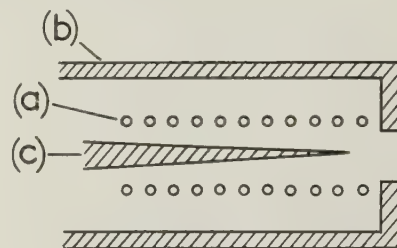


Fig. 7. Schematic diagram of proposed fluorescent X-ray tube

(a) Cathode; (b) Anti-cathode; (c) Molybdenum needle (filament potential) emitting  $\text{MoK}\alpha$  radiation.

It is clear that to obtain the desired increase of sensitivity, the intensity of the primary beam must be increased. In some cases the use of a primary beam defined by line slits instead of pinholes can give increased intensity.<sup>(32,33)</sup> However, if information on low-angle diffraction is required this method is not so suitable. It would be advantageous for many purposes to have a primary beam converging to a point and some attempts have been made to achieve such a beam.<sup>(35,36)</sup>

The effective brilliance of the source of X-rays for the chosen radiation (usually  $K\alpha$ ) cannot be increased much by increasing the voltage applied to the tube, but it may be possible to utilize monochromatic X-rays produced by fluorescence. In the arrangement tried by R. Griffon (Fig. 7) the source (a needle of molybdenum) is irradiated by an intense flux of X-rays produced by bombardment



of the surrounding anti-cathode. It is as yet too early to say what value this scheme will have.\*

Existing designs of moving-anode X-ray tubes enable a source of ten times the brilliance of an ordinary tube to be obtained quite easily but it does not seem to be easy to exceed this figure, due mainly to the rapid deterioration of target materials under these conditions. Even if this difficulty can be overcome, it will still be difficult to concentrate the increased tube currents on to a small focal spot due to the effect of the space charge in the tube. The use of a target of molten metal<sup>(23)</sup> has been suggested as a way of overcoming the first difficulty, but it seems likely that more can be achieved by increasing the efficiency of the water cooling of the anode. By providing increased cooling area, and by directing the water flow parallel to the surface to be cooled, a considerable increase in efficiency may be obtained. Although in these ways the brilliance of the tube may be increased by 50 or perhaps even 100, there seems to be no way of obtaining an improvement of 1 000 times or more.

For the accurate measurement of lattice parameters and line profiles, accurate monochromatization is necessary and, although crystal monochromators will separate  $\alpha_1$  and  $\alpha_2$  components, they cannot isolate narrower bands and it is difficult to separate the line broadening due to the specimen from that due to the spread of wavelengths in the source. It would seem to be better to try to secure achromatization for this purpose, and attempts have already been made to do this.<sup>(37)</sup>

Finally the availability of high-intensity sources opens up the possibility of quite new techniques. So far it has been possible to measure the amplitudes of the diffracted X-ray beams but not their phases, but recently methods have been suggested<sup>(38)</sup> for determining the phase differences between diffracted beams directly. For this and other new experiments in X-ray analysis, high-intensity sources and efficient utilization of the radiation are essential.

## MONOCHROMATORS AND COLLIMATORS

### General principles

DR. I. MACARTHUR opened the second session with consideration of "Some factors in precision monochromatization and collimation,"<sup>(39)</sup> showing their relation to tube design and their importance in modern problems. Filtering need not be confined to ordinary or balanced foils: failing smooth h.t., a considerable gain in the  $K\alpha$  content of a tube's output can be gained by mechanical or electrical shuttering (by grid control<sup>(40)</sup>) of beam production during the lower voltage intervals.

\* Later in the Conference Mr. W. J. Oosterkamp considered the use of radioactive material as a source of monochromatic K-radiation. For an output equivalent to that of an ordinary X-ray tube a source of 1500 curies would be required, but although this might be possible with artificial radioactive substances, such a source would have to have a half life of only 12 hours to give the required intensity.

An isolated sharp focus of fixed location and free of fine structure, such as obtains in the low-friction liquid-seal Leeds rotor<sup>(8)</sup> facilitates speed and interpretation, even without anterior defining slits. With the appropriate geometry, Soller slits can utilize large foci. Even careful setting of hemispherical cathode and point specimen, as in the low-wattage Hess tube<sup>(41)</sup> allows precise diffraction X-radiograms at moderate exposures despite spot extension to line hyperbola. The line focus at grazing side elevation is admirable for crystal monochromators. In gas tubes it can be approximated to by cathode moulding<sup>(42)</sup> or linear electron beam dispersion.<sup>(43)</sup> But its use in small-angle diffuse scattering involves correction<sup>(34)</sup> and may smear out important information.<sup>(44)</sup>

The characteristic features of double-stop slits, pin-hole and line, were illustrated from fibre X-radiograms. Precise secondary screening and avoidance of ordered-alloy stops is necessary in low-angle work. Factors involved in the economical use of stop slits<sup>(45)</sup> (geometrical, focal, nature and extent of specimen, resolution required) are forcing merited attention in the biological field.

Reflecting-wall collimators (calcite-faced line slits, lead-glass fine capillary tubes, converging polished walls<sup>(46)</sup>) yield a large gain in speed and some degree of monochromatization; admirable for wider-angle reflexions, even from large crystal cells,<sup>(47)</sup> they have to be used with caution in low-angle precision work owing to interference effects from micro-scale surface irregularities such as fatty layering, micro-corrosion, terracing or ripple.

In low-angle diffuse scattering studies with Dr. B. Patnaik, *pros* and *cons* had been investigated in the use of ground and elastically bent quartz plate monochromators. The transmission type,<sup>(48)</sup> of course (planes approximately radial), useful in analytical spectroscopy especially of artificial radioactive sources, suffers no refractive index correction and indeed gains additional focusing from the elastic lattice strain. Factors discussed quantitatively for the reflexion types used, included type and combination, geometrical errors of construction and setting, finite focus, refractive index correction, spectral purity, line width and contour, non-surface reflexion, screening or allowance for  $\alpha_2$  component, and especially crystal perfection, to which end Du Mond's adaptation of cycloid geometry to precision workshop practice<sup>(49)</sup> is notable. Major developments of the last three years which are setting up new technical standards include achromatic, point, and refractive focusing, and combinations of a wider range of geometrical forms. Dr. McArthur said they had been, or would be later, described by others.

### Focusing reflectors

In his paper on "The production of converging beams by total reflexion," DR. W. EHRENBURG described how the inner surface of a right elliptic cylinder may be used as an element in X-ray optics. Such a surface retains

its perfect focusing properties even when the angle  $\chi$  between the rays and the surface approaches zero.<sup>(50,51)</sup> X-rays are reflected only if  $\chi$  is smaller than the critical angle of total reflexion ( $\sim 10^{-3}$ ).

A suitable surface may be produced by bending an optical flat in a particular way<sup>(24)</sup> and calculations show that it is possible to construct the reflector sufficiently accurately to produce a beam of width  $1\mu$ . This arrangement gives 200 times the intensity of a slit system having the same definition and leaving the same working distance.

The possibility of producing fine focal lines of X-rays at a large distance from the optical system suggests that the total reflexion of X-rays can be used to investigate the structure of optical surfaces. It is found that when a convergent beam produced by total reflexion is used, some stray radiation is thrown into the neighbourhood of the focus, resulting in a widening of the X-ray image of the slit and the production of "wings."<sup>(52)</sup> Although air scattering, halation and Fresnel diffraction account for some of this widening, it may be shown that most of it is due to the action of the reflector. A bare glass surface shows the effect more than a surface coated with gold, but gold surfaces seem to deteriorate with time.

The nature of the focus obtained with polished surfaces suggests that the process of polishing leaves the surface in a state of imperfect flatness. This view is confirmed by the appearance of the reflexion from an artificially irregular surface produced by evaporating gold on to the reflector in such a way that the thickness of the coating varies sinusoidally along it. The results indicate that the polished surface consists of irregular hills and valleys of about 10 Å height and of width about 1 mm.

A focusing reflector of this type has two special properties. Firstly all hard radiation is suppressed in the reflected beam, and secondly the converging beam will damage a sensitive specimen less than a parallel or divergent beam since the specific X-ray dose received by the specimen depends on the ratio of X-ray intensity in the photographic emulsion to that in the specimen. In comparing the intensity obtained by the reflector to that with a slit system it is necessary to distinguish two cases.

For back reflexion work and other work where the wings in the image are not important it may be shown that the ratio  $R$  of the intensity obtained with reflector to that with the comparable double slit is given by

$$R \approx 0.012 L/W$$

where  $L$  is the free distance required in front of the specimen, and  $W$  is the width of the focus. Thus a small focal spot is necessary if the reflector is to be worth while.

On the other hand, if weak interferences are to be observed in the neighbourhood of the geometric focus it will be necessary to suppress the wings by the use of an additional slit between plate and reflector and the resulting width of the direct beam will be, for long exposures, the pinhole image by this wing slit of the used

portion of the reflector. It may be shown that in this case

$$\begin{aligned} R &\approx 2 && \text{for } \theta < 0.006 \\ R &\approx 0.012 && \text{for } \theta > 0.006 \end{aligned}$$

where  $\theta$  is the minimum angle of deflexion resolved.

Hence for small values of  $\theta$  the optical method gives twice the intensity in addition to an improvement in spot definition and the reduction of Compton scattering.  $R = 1$  for  $\theta = 0.012$ , corresponding to a lattice spacing of about 100 Å. For shorter spacings the use of ordinary slits is preferable.

#### Monochromators.

MR. P. M. DE WOLFF (Technisch Physische Dienst Delft) described work by himself, Mr. T. J. Tiedema (Delft) and Mlle Cauchois (Paris) on monochromators.

Mr. Tiedema has worked out a technique of growing large single crystals of aluminium for use in curved crystal monochromators. These show much less extinction than the usual quartz or mica and therefore yield a much higher reflected intensity. Crystals of the correct shape and with the lattice planes appropriately bent can be grown by making the crystal grow into the bent metal strip.<sup>(53,54)</sup> After annealing the resolving power is comparable with that of a quartz monochromator but the reflected intensity of rays passing through the crystal is roughly ten times greater and gives very good results in the study of spectra below 2 Å.<sup>(55)</sup>

Mr. de Wolff described his own work on the construction of monochromators using bent plane parallel quartz slabs. By bending to a logarithmic spiral form a very substantial reduction of the extent of the aberrations may be achieved.<sup>(56)</sup> A careful kinematic design ensures exact equality of the bending torques at the top and bottom of the crystal and, to avoid large local stresses, the forces are exerted on tiny brass cylinders glued on to the quartz.

This monochromator is used with a multiple powder diffraction camera based on the focusing principle used by Guinier.<sup>(57)</sup> Such a camera is at least as economical as a common powder camera without monochromator and, of course, the gain in information is considerable except for back-reflexion lines. However, it is clear that the low angle lines are always the most characteristic and this is even more true with a high resolving power and increased precision. Once the correct elementary cell has been found, there is no doubt as to the indexing of the lines even if only 10–20 lines have been used because practically every possible reflexion is found well resolved.

MR. P. GAY (Cavendish Laboratory, Cambridge) described "A parallel beam concentrating monochromator."<sup>(58)</sup> An enhancement of the intensity reflected from such a monochromator is obtained by the geometrical foreshortening effect produced by reflexion from a surface inclined at an angle  $\phi$  to the Bragg planes and the reflected intensity has been studied as a function of  $\phi$  and of the treatment of the crystal surface.

It can be shown<sup>(59)</sup> that the reflected intensity from an



deal mosaic should increase by a factor of two as  $\phi$  changes from 0 (symmetric position) to  $\theta$  (concentrating position). More recently it has been shown that this ratio may be greater than two.<sup>(60)</sup> By grinding the crystal at an angle to the Bragg planes and then rotating it about the normal to the reflecting planes, a range of values of  $\phi$  can be produced on the same crystal surface. The experimental results (Fig. 8) may be explained by the presence of an absorbing layer on the surface of the crystal. In this case, when the angle of grinding is equal to the Bragg angle, the reflected beam never emerges from the layer.

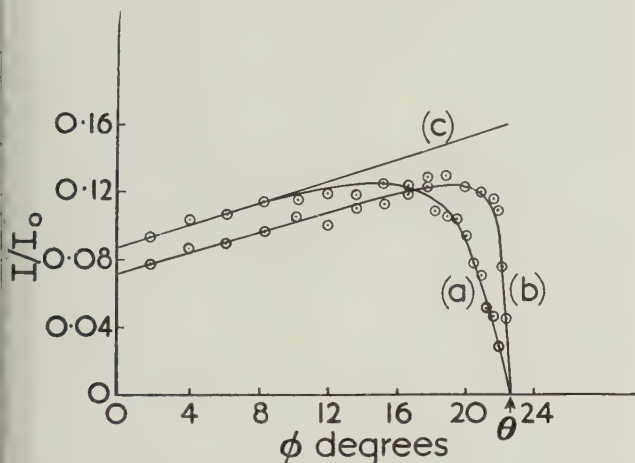


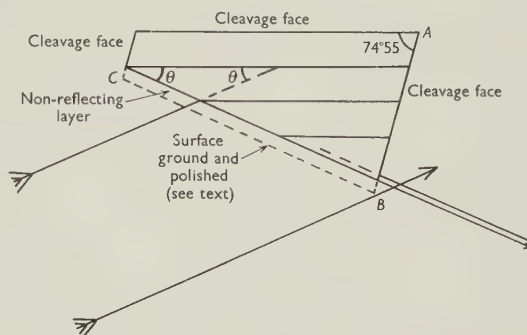
Fig. 8. Variation of  $I/I_0$  with  $\phi$  for reflexion from a lithium fluoride crystal: (a) ground crystal; (b) same crystal after etching; (c) curve predicted by mosaic theory for a crystal without an abraded layer

The rapid decrease of the intensity of the reflected beam as  $\phi$  tends to  $\theta$  leads to the idea of the "cut-off" crystal (Fig. 9) when a fine beam of maximum intensity is required. The reflected beam emerges from the face  $AB$  without traversing any great path length in the absorbing layer so that the full two-fold intensity improvement would be expected.

Results for crystals of lithium fluoride show that relatively light grinding produces a marked increase in the reflected intensity. This behaviour can be interpreted in the following way. Light abrasion produces misorientations within the range of divergence of the incident beam so that all the misoriented material can reflect simultaneously whilst heavy working of the surface produces material of such large misorientation that it cannot reflect the incident beam.

For best results with the concentrating monochromator the crystal should be ground at the angle corresponding to the required width of beam, but should not exceed that value at which the intensity is a maximum. (By reversing the direction of the beams it is possible to produce very wide uniform beams—up to 4 mm in width). After grinding, the crystal should be etched to remove as much of the absorbing material as possible, and if high resolution is required the crystal should be

used in the etched state or alternatively a cleavage face used (2022 reflexion from calcite gives excellent results). For a high intensity of reflected beam the best results have been obtained with lithium fluoride crystals ground to the appropriate angle, etched, and then ground again lightly on fine carborundum.



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Fig. 9. "Cut-off" concentrating monochromator of calcite

DR. W. H. TAYLOR described work on the "Use of monochromatic radiation in alloy researches" carried out by DR. M. HARGREAVES.<sup>(61)</sup> Using  $\text{FeK}\alpha$  radiation reflected from a bent crystal of synthetic lithium fluoride and a camera arrangement such that only the  $\alpha_1$  beam falls on the specimen, it has been possible to obtain very clear photographs of the X-ray diffraction pattern from alloys with a composition near to  $\text{Cu}_4\text{FeNi}$ . These alloys, as has been shown before,<sup>(62,63,64,65)</sup> are single phase after quenching from high temperatures and, after long annealing, consist of lamellae of two tetragonal structures. After a very short anneal the alloy can be brought into an intermediate state: this "modulated" structure shows an X-ray powder pattern in which each line is accompanied by "side-bands."

The improved accuracy of measurement has led to a proposed modification of the original theoretical discussion. It now appears that a nucleation-and-growth hypothesis leads, without difficulty, to a quantitative explanation of the intensities of the side-bands relative to the main line, and of the asymmetry of the two side bands on either side of a given line.

#### APPLICATION OF HIGH-INTENSITY X-RAY BEAMS

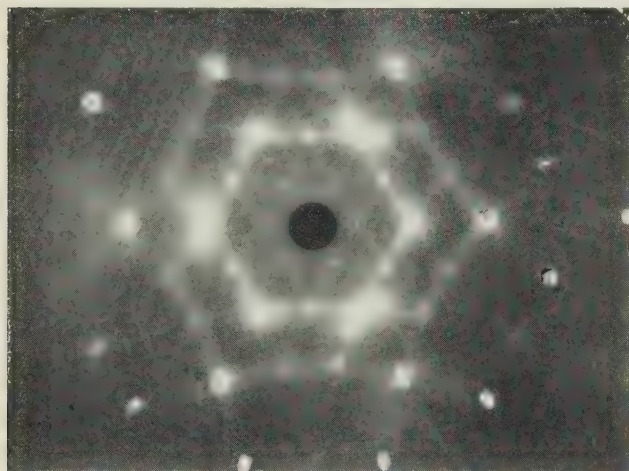
Fine clean powerful X-ray beams may be either essential or expedient. They are essential in the study of fleeting changes, unstable specimens, or in diagnostic control of critical industrial processes. Expediency varies in importance. DR. D. P. RILEY showed their value in his paper "The application of high-intensity X-ray beams in biological and colloid chemistry," choosing three typical examples of the study of weakly diffracting liquid disperse systems.

(i) Studies with J. H. Schulman and T. S. MacRoberts<sup>(66)</sup> of non-birefringent oil-soap-water systems titrated to transparency and fluidity by adding aliphatic or alicyclic alcohols, revealed definite low-angle diffraction bands.

Their spacings support a structure of close-packed uniform water spheres in oil, or oil spheres in water, each droplet being stabilized by a mixed monolayer of soap and alcohol molecules. By altering the concentration ratio of the constituents, droplet diameters and mean inter-distances could be made to vary between  $\sim 100$ – $500$  Å.

(ii) Studies with D. Herbert<sup>(67)</sup> of concentrated protein solutions gave a number of clearly defined bands yielding useful information as to size, shape, hydration, and nature of aggregation of the molecules. Human haemoglobin, egg albumin, and bovine serum albumin have been examined.

(iii) Studies with G. Oster were being made of sodium desoxyribonucleate (from thymus) and its equivalent nucleoprotein. The pseudolattice of these paracrystalline solutions expands on dilution, and molecular swelling is also evidenced. The variation of both low- and medium-angle diffraction detail with concentration and origin of specimen was being closely examined because of the importance of nucleic acid derivatives in living cells.



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Fig. 10. Diffuse scattering from benzil  
(copper  $K\alpha$  radiation)

DR. P. G. OWSTON (University College, London) described some results on the "Measurement of diffuse scattering and scattering by hydrogen atoms." Diffuse scattering<sup>(68,69,70)</sup> is a second order effect and a diffuse spot with an intensity 1% of that of the corresponding Bragg reflexion is considered very strong so that, using ordinary X-ray tubes, the exposure time is about one hour even for soft crystals. Since a very large number of exposures are required to map out the outlines of the diffuse scattering, even the preliminary investigation takes several weeks. When more detail is required, cleaner radiation is necessary and a moving-anode tube is useful because the effect of deposition of tungsten from the target is less serious. A direct current supply enables the optimum ratio of white to characteristic radiation to be maintained but even so a monochromator is usually necessary and this of course involves a

large increase in exposure time. Thus the use of a high intensity X-ray tube means not only greater convenience but also enables more useful information to be obtained.

The extra scattering is not all temperature dependent and it would be of interest to study the vibrations causing the temperature dependent scattering at a transition point. It is usually supposed that the vibrations then become so powerful that the lattice forces are overcome but it is not known if this applies to all vibrations. Very accurate temperature control would be necessary to work near a transition point so that it is essential to shorten the exposure time. Another problem which can be attacked only by increasing the intensity of the incident X-ray beam is the explanation of the background of the diffuse scattering pattern. Such a pattern consists of diffuse spots, which can be fairly well explained in terms of the elastic properties of the crystal, joined by diffuse streaks which are less well understood (Fig. 10). Unfortunately these are very weak and often approach the cosmic ray background when an ordinary X-ray tube is used and under these conditions they are impossible to measure accurately.

In simple structures where the atomic or molecular centres lie in special positions, certain reflexions can be calculated to be absent, over and above the absence due to the symmetry of the space group, e.g. in ice all reflections with  $l = 4$  should be absent.<sup>(71,72,73)</sup> However, careful experiments show that these reflexions are not all absent but they are usually very weak. Their appearance can be due only to a lack of spherical symmetry of the water molecule, presumably due to the electrons in the O-H bond. The importance of O-H bonds makes an accurate study of the electron distribution of the greatest interest. The situation calls for a high-intensity tube which would enable Weissenberg photographs to be obtained in one day and thus enable reliable measurements to be made.

#### *The Structure of Cold-Worked Metals.*

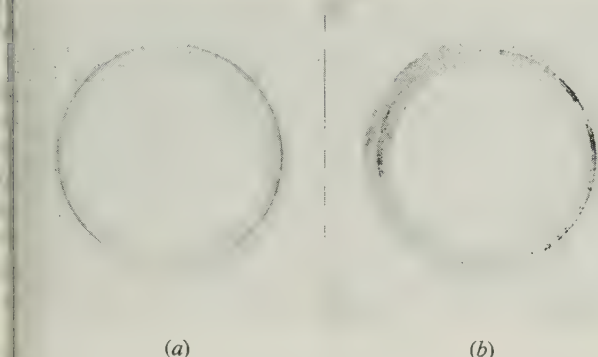
X-ray line broadening may be interpreted in two ways—strain broadening<sup>(74)</sup> or particle size broadening.<sup>(75)</sup> Different results have been obtained by different workers and from the evidence it appears that both effects are present. However, if the material consists of small discrete particles, then by irradiating only a small number of these, the diffraction ring should show spots as with annealed material. Mr. P. B. HIRSCH, speaking on "A microbeam technique applied to the study of cold-worked metals," described the results obtained in this way.<sup>(76)</sup>

A specimen of pure aluminium of grain size  $40 \mu$  was reduced in thickness 60% by cold-rolling. After etching, back reflexion photographs were taken with 1 mm and with  $35 \mu$  diameter collimators (Fig. 11). These show clearly that the cold-worked metal consists of sharply defined regions misoriented from each other (particles). The mean grain volume of the particles can be obtained from the number of spots on the ring.<sup>(77)</sup>



for the material examined soon after rolling an approximate value of the particle size is  $3.6 \mu$  whereas one year after rolling the mean particle size is found to be about  $1 \mu$ . Thus it seems that the particle size decreases slightly with time.

More information about the causes of line broadening and the physical nature of the particles may be obtained from an examination of the shapes of the spots, in which both a tangential and a radial breadth can be distinguished.



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Fig. 11. Back reflexion photographs from cold-rolled aluminium: (a) 1 mm collimator; (b)  $35 \mu$  collimator

ished. Results show that both these quantities decrease with time after rolling, corresponding to the process called recovery, and it is clear that the line breadth must be due partly to the breadth of the spots, i.e. either to the shape of the particles or to a range of strains within them, although part of the line breadth may be due to possible mean strains of each particle giving rise to an "off-line" of the spot as a whole.

The radial breadth of the spots for the material examined a year after rolling is found to be about  $0.8 \times 10^{-3}$  radian. If the material consisted of equiaxed undistorted particles their size determined from this breadth would be  $0.15 \mu$ , whereas from the method of counting spots it is found to be  $2 \mu$ . For the material examined soon after rolling the discrepancy is very much greater. However, the particles may be non-equiaxed, such as lamellae or rods, and give rise to appreciable broadening due to small size in some directions. Such shapes of particle would give rise to spots whose axes were, in general, neither tangential nor radial and spots of this type do occur frequently. The radial and tangential breadths of the spots would, in the case of no strain broadening, give roughly the dimensions of the particle in two perpendicular directions and since the volume of the particle is known, the third dimension may be calculated. For the material examined soon after rolling the third length would be several millimetres and it must be concluded that the broadenings cannot be explained in terms of particle shape alone.

The large breadths of the spots soon after rolling are mostly due to distortion. If due entirely to this cause the breadth can be explained if the particles are bent

over an arc of about  $1.5 \times 10^{-3}$  radian, corresponding to a range of stresses of the order of the measured yield stress for this material ( $10^9$  dynes/cm<sup>2</sup>). It is not yet known how much of the broadening of the spots for the material after recovery is due to distortion and how much to particle size.

For very small deformations the original crystal gives a series of spots close together and covering a large range of angles on the ring. Sometimes these spots have their axes neither along nor perpendicular to the ring and are parallel to each other such as would be expected for a series of slip lamellae all slightly misoriented relative to each other, and it is thought therefore, that the polycrystalline material deforms initially by slip. On further deformation the spots are no longer parallel to each other probably due to slip on other planes or to rotation.

If the material is examined immediately after rolling some of the spots are very broad and have very long tails extending far off the ring. On recovery these tails disappear, the main broadening and the background are reduced, and the particle size decreases. The interpretation of this is that the strains are relieved by polygonization,<sup>(78)</sup> a process by which one bent particle splits up into a number of unbent particles of slightly different orientations.

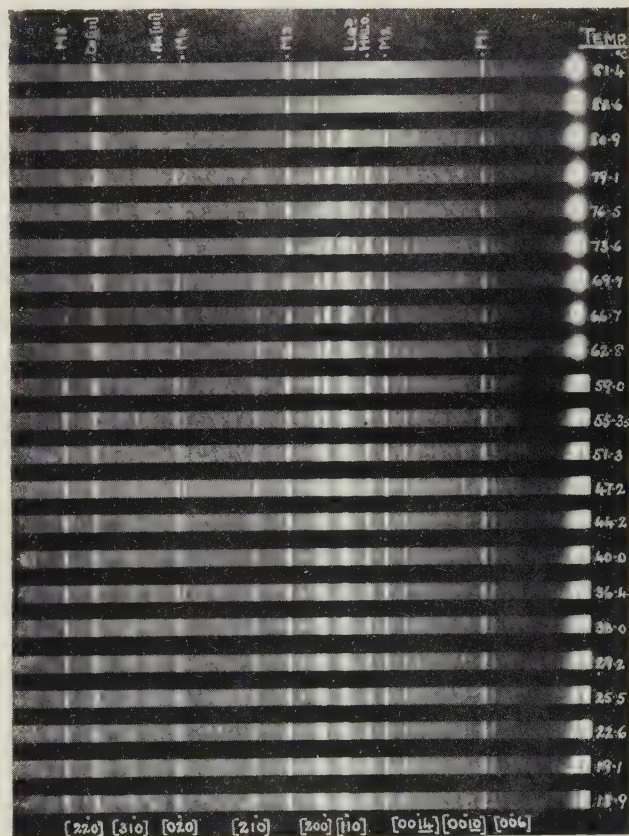


Fig. 12. Powder X-radiograms of dipalmityl ketone between room temperature and melting point ( $\times \frac{3}{4}$ )

R  $\sim 12$  cm., mica calibration. 5 min exposures. 5 kW generator, Davy Faraday Laboratory, Royal Institution, London.

Taking as his application a member of the time-expedient class, DR. I. MACARTHUR in "Temperature-variation of structures in homologous series" sketched the advantages gained by a comparative study of homologues under varied conditions. Following Müller,<sup>(79)</sup> and with the valued assistance of Mr. H. Smith, the 5 kW X-ray generator of the Davy Faraday Laboratory had been used to record powder photographs of *n*-aliphatic long-chain compounds at intervals between room temperature and melting point. Five-minute exposures (with 5-minute inter-adjustments) were given at 10–15 cm distance on a drum-type camera providing a sequence of equatorial exposures on a half-plate film. Temperature control by a circulatory system incorporating reservoir, heater and centrifugal pump proved accurate to 0.2° C.

Results, which include low- and high-energy discontinuous transitions and continuous anisotropic variation (Fig. 12), were presented for a number of series, with chain length, polar group, position of polar group, and temperature as parameters. The results, interpretable in force-field terms, were especially significant in the light of complementary physico-chemical structure-analytical techniques, especially thermal and dielectric measurements.

MR. P. GAY discussed some results obtained in a "Study of the abrasion of crystals." The most uniform portion of the X-ray beam reflected from a parallel beam monochromator crystal is allowed to fall on to the ground surface of a calcite crystal. As the divergence of the beam is small, only material which is misoriented from the matrix by less than the divergence of the beam can reflect, and thus small misorientations of the material in the worked surface give large reflected intensities. It is found that when the incident X-ray beam strikes the crystal at grazing incidence, so that the surface layer is illuminated, the diffraction rings show the presence on the surface of a randomly oriented powder. Even after brushing and washing the crystal, relatively large misorientations (of the order of 25° in calcite) still remain.

The thickness of the layer of misoriented material is measured by investigating the reflected intensity as a function of  $\phi$ , the angle between the reflecting planes and the surface. A beam reflected from the matrix cannot be reflected from the misoriented material which acts as an absorber. The ratio of reflected to incident intensity ( $I/I_0$ ) is given by

$$\frac{I}{I_0} = \frac{K}{\mu(1+R)} \exp \{ -\mu' t [\operatorname{cosec}(\theta + \phi) + \operatorname{cosec}(\theta - \phi)] \} \quad (1)$$

where  $\mu$  = linear absorption coefficient of matrix.

$\mu'$  = linear absorption coefficient of layer.

$t$  = thickness of layer.

$K$  = volume reflexion coefficient of matrix.

$R$  = concentrating ratio.

and a typical experimental result is shown in Fig. 8. The parameter  $\mu' t$  in the theoretical formula is adjusted

to give the best fit of the experimental points. It is a measure of the amount of absorbing material in the layer but, as the density of this material may be less than that of the matrix, the thickness  $t$  obtained by putting  $\mu = \mu'$  in this parameter may be less than the actual thickness of the layer. The thicknesses obtained in this way vary from 1–10  $\mu$ .

Equation (1) assumes a sharp discontinuity in the reflexion coefficient at the matrix-layer interface. This is unlikely to be the case, the effect on the reflected intensity of various distributions of reflexion coefficients has been investigated theoretically and the following physical picture is obtained. Deep within the crystal the deformation varies slowly with distance and the reflexion coefficient is approximately constant. As the surface is approached the deformation—and hence the misorientation—increases, and at the point at which the density of the material begins to decrease the misorientations begin to increase more rapidly. Finally, at the surface most of the material is so widely misoriented that it cannot reflect at all.

The values of the intensity ratio in the symmetrical position ( $\phi = 0$ ) vary widely with the surface treatment and it follows that the deformation takes place within the crystal to within the depth of penetration of the beam. It is clear from the experiments that distortions of very large angular range and great depth are caused during abrasion.

It is possible that the method described above can be used for the non-destructive measurement of the thickness of a metallic film deposited on a polycrystalline base.

#### *Discussion on application of high-intensity X-ray beams*

In the discussion which followed, MR. J. S. THOM described a microbeam technique for studying the mosaic structure of crystals in which the crystal surface is scanned by a beam of diameter 70  $\mu$  and divergence about 9' of arc. Results on a polished cleavage face of calcite indicate that there are no relative misorientations of more than about 2' of arc and support the hypothesis of recrystallization on the cleavage face after grinding.

DR. H. WILMAN (Imperial College, London) described results obtained from an electron diffraction study of unidirectionally abraded crystal surfaces. He suggested that these results showed that a new kind of slip mechanism is operative—rotational slip.<sup>(80)</sup>

PROF. GUINIER suggested that microbeam transmission photographs of thin foil would be rather easier to interpret than the back reflexion photographs described by Mr. Hirsch.

#### TECHNIQUES OF X-RAY ANALYSIS

One short session of the Conference was devoted to a number of small items of X-ray technique. MR. FOURNET described how diffraction bands corresponding to a spacing of 240 Å can be observed by the use of two bent-crystal monochromators in series to suppress parasitic scattering, and MR. E. G. STEWARD referred



o simple modifications to standard cameras to enable diffraction patterns at high or low temperatures to be recorded.<sup>(81)</sup>

Some problems encountered in microbeam work, including microbeam collimation and the directional properties of Geiger counters were considered by Mr. P. B. HIRSCH and Mr. J. S. THORP. The latter showed that quite large changes in angular setting of the counter produce only small changes in recorded intensity provided that the appropriate region of the sensitive volume of the counter is chosen.

DR. T. LL. RICHARDS demonstrated a dynamic model illustrating structural effects in face-centred cubic metals, and Mr. C. J. LEES showed a simple electronic stabilizer for X-ray tube currents. Other subjects discussed were the preparation of thin windows of beryllium foil,<sup>(82)</sup> a simple scanning mechanism for use with large-grained polycrystalline specimens,<sup>(83)</sup> and the use of Geiger counters for the measurement of integrated reflexions.<sup>(84)</sup>

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## ORIGINAL CONTRIBUTIONS

### The spectral emittance of nickel- and oxide-coated nickel cathodes

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The spectral emittance values at a wavelength  $\lambda = 0.66 \mu$  have been measured for various types of oxide-coated cathode, and for nickel cores, using a cylindrical diffuse reflectometer. The influence of various factors, essentially affecting the surface condition, on the emittance has been examined. Tables show for nickel the effect of stoving treatment, temperature, and getter flash; and for oxide-coated cathodes the strong effect of the oxide surface texture, and the subsidiary effects of glass tube baking temperature, and getter flash.

The thermionic emission from a cathode varies exponentially with the cathode temperature, and it is, therefore, essential that the temperature should be measured as accurately as possible in any systematic study of emission. The most satisfactory method of determining the cathode temperature, considering both accuracy and practical application in commercial valves, is to measure the brightness temperature with an optical pyrometer of the disappearing filament type. To obtain the true temperature from the measured temperature the spectral emittance†  $e_\lambda$  of the cathode surface must be known.

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† The terminology suggested by Worthing<sup>(1)</sup> has been adopted in this paper. Terms ending "ivity" refer to properties inherent to the material measured under specific conditions and independent of the surface. Where measurements apply to a particular surface state the ending "ance" is used.

Published data on the spectral emittance of cathodes used in thermionic valves are sparse, and the values quoted differ widely, either because of the difference in the method of measurement or more probably because of variations in the surface condition of the cathodes investigated.

For oxide-coated cathodes Champeix<sup>(2)</sup> reported that the spectral emittance decreased exponentially with coating thickness, and that for the same thickness the value for spray-coated cathodes differed from that of cathaphoretically-coated cathodes. This dependence of emittance on the method of coating was also reported by Moore and Allison,<sup>(3)</sup> and might be expected since the method of coating affects the surface texture. Further, Champeix<sup>(2)</sup> found that the spectral emittance of coated cathodes did not vary significantly with temperature, whereas for the nickel core it increased when



the temperature was raised from room temperature to 377° C. Entirely similar results had been indicated in earlier work by Rees and Munday<sup>(4)</sup> in these laboratories, for etched nickel cathodes and cataphoretically-coated cathodes.

In the present work the influence of other factors which may either affect the true emittance or the experimental determination of emittance have been examined, both for uncoated nickel cathodes and nickel cores coated with barium strontium oxide. The investigations on nickel cathodes cover the variation of emittance with stoving treatment and temperature, and the effect of the getter used in the tube. Those for oxide-coated cathodes include the effect of temperature used for degassing the glass envelope, getter contamination, and the surface texture of the coating.

### THEORETICAL

The emittance values were computed from measurements of reflectance made in a diffuse reflectometer similar to that used by Prescott and Morrison.<sup>(5)</sup> This method was chosen since it eliminates errors due to the thermal conductivity of the coating associated with black body measurements, and to the effect of local cooling associated with the use of spots of known emissivity or with thermocouple probes.<sup>(6)</sup>

The principle is to illuminate the cathode with diffuse homogeneous light from all directions, and to measure the proportion of light reflected by the cathode in one direction, normal to the cathode surface. Since this is essentially the same as illuminating in one direction and measuring the amount of light reflected in all directions the hemispherical spectral reflectance  $r_{\lambda h}$  at wavelength  $\lambda$  at which measurements are made is obtained. The hemispherical spectral emittance  $e_{\lambda h}$  is computed from  $r_{\lambda h}$  according to Kirchhoff's law:

$$e_{\lambda h} + r_{\lambda h} = 1$$

In calculating the true cathode temperature from the brightness temperature, the normal spectral emittance  $e_{\lambda n}$  should be used. The two emittance values differ on account of the departure from Lambert's cosine law exhibited by the radiation from non-black bodies. In practice, however, this departure is small, amounting to a temperature variation of a few degrees per mm across the heated cathode, and thus the hemispherical spectral emittance can be used in computing true from brightness temperature.

The diffuse illumination is obtained by reflexion from the walls of a large cylinder, in which the cathode is mounted, and which is lit by lamps. It is, therefore, essential for homogeneity that the walls should be covered with a diffuse material of high reflectivity, that no direct light from the lamps should fall on to the cathode, and that the cathode should be mounted concentrically in the cylinder. The cathode and background are viewed through a small hole in the cylinder and the

apparent brightness temperature of the cathode compared with that of the background using a disappearing filament optical pyrometer.

The formula for calculating the spectral reflectance was derived by Prescott<sup>(7)</sup> and is given by

$$r_{\lambda} = \exp \left[ -C_2/\lambda(1/T_1 - 1/T_2) \right] - \exp \left[ -C_2/\lambda(1/T_3 - 1/T_2) \right] \quad (1)$$

where  $C_2$  = radiation constant = 1.438 cm deg.

$\lambda$  = wavelength at which measurements are made.

$T_1$  = brightness temperature of the cathode when illuminated in ° K.

$T_2$  = brightness temperature of the background.

$T_3$  = brightness temperature of the cathode when lights in the reflectometer are turned off.

The second term in the equation will be zero when the cathode is at room temperature. Since the cathodes were mounted in glass tubes a correction to the measured emittance had to be applied to allow for absorption in, and reflexion by the glass.

The correction is derived by considerations similar to those used by Hamaker.<sup>(8)</sup> If the intensity of the incident light of wavelength  $\lambda$  in the reflectometer is  $I$ , then the intensity of the background viewed through the tube is

$$I_B = I[t_g^2/(1 - r_g) + r_g] \quad (2)$$

where  $t_g$  and  $r_g$  are respectively the transmission and reflexion factors of the glass at wavelength  $\lambda$ . The intensity of the cathode seen through the glass tube will be

$$I_c = I[r_g + r_{\lambda}t_g^2/(1 - r_g)] \quad (3)$$

where  $r_{\lambda}$  = the true reflectance value for the cathode surface. Therefore, since the measured reflectance  $r'_{\lambda}$  is given by  $r'_{\lambda} = I_c/I_B$  and  $e_{\lambda} + r_{\lambda} = 1$ , it follows that the true emittance  $e_{\lambda}$  is related to the measured value of  $e'_{\lambda}$  by

$$e_{\lambda} = e'_{\lambda}[1 + r_g(1 - r_g)/t_g^2] \quad (4)$$

The relation between  $r_g$  and  $t_g$  can be determined experimentally in the reflectometer by measuring the brightness temperature of the background with and without the glass tube present.

By Wien's law,

$$I_B/I = \exp \left[ -C_2/\lambda(1/T_4 - 1/T_5) \right] \quad (5)$$

where  $T_4$  = apparent temperature of background through the glass tube.

$T_5$  = apparent temperature of background without the tube present.

Then from (2)

$$r_g + t_g^2/(1 - r_g) = \exp \left[ -C_2/\lambda(1/T_4 - 1/T_5) \right] \quad (6)$$

Thus only  $r_g$  or  $t_g$  need be determined separately.

## EXPERIMENTAL

(a) *The Experimental Tubes.* The nickel cathodes were of grade "O" quality by Henry Wiggin and Co. Ltd. (specification: Ni + Co > 99.5%, total impurities < 0.7% with: C < 0.04%, Fe < 0.2%, Mn < 0.15%, S < 0.005%, Mg 0.07–0.15%, Si < 0.1%, Co < 0.5%, Cu < 0.1%), 2 mm diameter and 2 cm long, heated indirectly by alundum-covered spiral tungsten heaters. The cathodes were electrically polished in 1:1 sulphuric acid, washed, and stoved in hydrogen at 900°C for 5 min before assembly. In certain cases there was an additional treatment of vacuum stoving at 1030–1080°C. For the coated nickel cathodes the cores, 1.8 mm diameter, 2 cm long, were etched to a matt surface and stoved in hydrogen as before. The majority were coated by spraying with a suspension of approximately equimolar barium strontium carbonate (precipitated with sodium carbonate from a solution of the nitrates, washed and dried at 650°C) in methyl alcohol with a nitrocellulose binder. Some coatings were cathodetically applied from suspensions of ammonium precipitated carbonates; these include smooth coatings on nickel, strontium oxide and barium strontium oxide on molybdenum strip, and double and triple oxide on tungsten filaments. The coating thickness was measured with a Leitz projection microscope. The smooth glossy coatings made up for experimental purposes were obtained from colloidal suspensions prepared by ball milling the carbonates for long periods in ethylene glycol and suitably diluting with methyl alcohol according to a recent patent.<sup>(9)</sup>

The cathodes were mounted in tubes with a getter assembly, either a barium-filled iron tube or a zirconium wire,<sup>(10)</sup> screened from the cathode by a mica shield. In certain cases barium-magnesium getters were used. These consisted of pellets mounted in nickel boats and were not screened by mica. The tubes were pumped with a mercury diffusion pump backed by a rotary pump to a pressure of about  $10^{-5}$  mm mercury and the envelopes baked out at 400°C for 1 h on the pumps, unless otherwise stated. The nickel cathodes were then outgassed at 850°C brightness temperature and the coated cathodes formed and heat-activated at approximately 950°C. The getters were then activated and the tubes sealed off.

(b) *The Reflectometer.* The reflectometer consisted of a metal cylinder *A* (Fig. 1) 50 cm long and 14 cm diameter, coated on the inside with a matt magnesium oxide paint, and with a viewing hole *B*, 1 cm diameter, located centrally. Two end caps *C* each carrying a 12 V 48 W bulb, mounted centrally, fitted tightly on the ends of the cylinder. The tops of the bulbs were painted white so that no direct light fell on the cathode under investigation. The tube containing the cathode was held by a clip *D* which could be adjusted in a vertical plane. The cathode could be heated by two leads through the top cap.

The homogeneity of the light was tested by suspending

a V-shaped piece of white paper centrally at various angles with the fold towards the hole, and comparing the apparent temperatures of the paper on each side of the fold. In no case was the difference more than 2 or 3°, and thus the light was considered sufficiently homogeneous for the purpose of the experiments. It had been thought that where a getter was evaporated in the tube the black deposit on the glass might cause

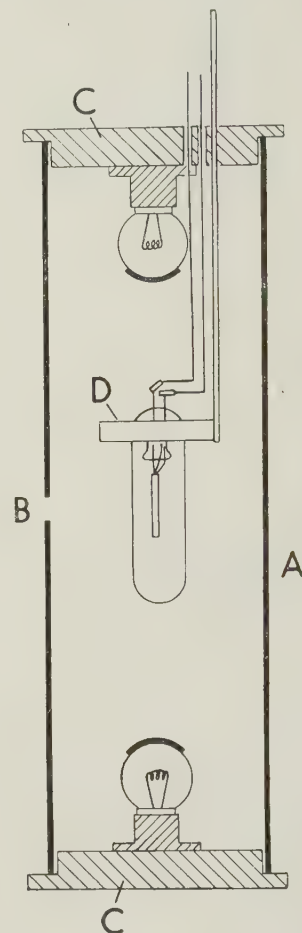


Fig. 1. The reflectometer

some inhomogeneity in the light distribution. To investigate this point a number of tubes were constructed containing both a barium and a zirconium getter. Both getters were outgassed on the pumps and the tubes sealed off. The zirconium wire was then heated to ensure a good vacuum and the emittance determined. The top of the tube was then painted black over an area corresponding to that normally occupied by the barium getter film, and the emittance again measured. The barium getter could then still be fired when investigating the influence of the getter. The results in Table 1 showed that blackening the glass tube had no significant effect on the emittance values obtained.

The reflexion and transmission factors of the glass



Table 1. Effect of getter flash and bulb baking temperature on spectral emittance at room temperature of oxide-coated cathodes (sprayed; Na-pptd. Ba/Sr carbonate). Coating thickness  $60\mu$ . Mean values and standard deviation  $\sigma$  for  $N$  cathodes.  $\lambda = 0.66\mu$ 

Series No.	Getter	Bulb Baking Temperature											
		$N$	$300^{\circ}\text{C}$ $e_{\lambda}$	$\sigma$	$N$	$350^{\circ}\text{C}$ $e_{\lambda}$	$\sigma$	$N$	$400^{\circ}\text{C}$ $e_{\lambda}$	$\sigma$	$N$	Overall mean $e_{\lambda}$	$\sigma$
1	Zr and Ba getter in same tube—												
	(a) Zr alone used	4	0.14(8)	0.036	3	0.12(4)	0.024	3	0.18(3)	0.024	10	0.15	0.039
	(b) Bulb blackened	4	0.17(4)	0.045	3	0.11(6)	0.015	3	0.21	0.026	10	0.16(5)	0.049
	(c) Ba getter fired	4	0.16(5)	0.035	3	0.18(5)	0.022	3	0.22(5)	0.04	10	0.18(9)	0.042
2	Ba getter alone	2	0.13	0.02	—	—	—	4	0.16	0.05	6	0.14(7)	0.033
3	Ba-Mg getter (unscreened)	—	—	—	4	0.24	0.082	1	0.22	—	5	0.23(5)	0.08
	Mean of 1(a) and 1(b)	$4 \times 2$	0.16	0.042	$3 \times 2$	0.12	0.02	$3 \times 2$	0.19(7)	0.032	$10 \times 2$	0.15(9)	0.047
	Mean of 1(c) and 2	6	0.15(2)	0.035	3	0.18(5)	0.022	7	0.19	0.063	16	0.17(5)	0.033
	Mean of 1 and 2	$4 \times 3$ +2	0.15(7)	0.039	$3 \times 3$	0.13(2)	0.021	$3 \times 3$ +4	0.19(4)	0.05	$10 \times 3$ +6	0.16(5)	0.041

envelopes were both measured directly, and the reflectometer method of obtaining the relation between them (equation 6) used as a check.  $r_g$  was determined by focusing a monochromatic beam on a slit  $1\text{ cm} \times 2\text{ mm}$  in a black paper cover wrapped tightly round the glass tube, the reflected beam being measured photo-electrically.  $t_g$  was measured by directing a monochromatic beam through the tube as near the centre as possible. The mean values thus determined on six tubes containing various cathodes were  $t_g = 0.86$  and  $r_g = 0.08$  (for  $\lambda = 0.66\mu$ ) with a maximum spread of 10%. Reflectometer measurements of  $T_4$  and  $T_5$  also made on six tubes gave a mean value of  $0.82 \pm 0.03$  for the exponential term in equation (5). With this value and  $r_g = 0.08$ , equation (6) gives  $t_g = 0.82$  in fair agreement with direct measurement. Since a 10% error in either  $r_g$  or  $t_g$  results in only a 1% error in the correction factor (equation 4) the values of  $r_g = 0.08$  and  $t_g = 0.82$  were used for all tubes. Substituting these values in equation (4),

$$e_\lambda = 1.11e'_\lambda \quad (7)$$

As a check, values of  $T_4$  and  $T_5$  were taken for each tube. These were consistent for all tubes within the limits of experimental error.

In general, emittance values were determined for three different points on the cathode surface and the mean taken as characteristic of that cathode. The standard deviation for most cathodes was 0.03, although the spread in the case of coated cathodes was sometimes as much as  $\pm 0.06$ . Measurements were made on several cathodes under each set of conditions and the mean for the batch determined. In assessing the effects of the various factors a "Students  $t$ " significance test was used.<sup>(11)</sup> In certain doubtful cases the test was applied to all the readings taken and not simply to the mean for individual cathodes. A disappearing filament optical pyrometer by the Cambridge Instrument Co., Ltd., fitted with a red filter of effective wavelength  $0.66\mu$ , was used for determining brightness temperature, the accuracy of measuring temperature differences being  $\pm 3^\circ\text{C}$ . This would make a corresponding error in emittance of

$\pm 0.025$ . In computing the true temperature from the brightness temperature, this will result in an error of  $\pm 4^\circ$  at  $1100^\circ\text{K}$  for an emittance value of the order of 0.3, but for an emittance of 0.1 the error will be about  $15^\circ$ .

## RESULTS

**Nickel Cathodes.** For the uncoated nickel cathodes the effects on the emittance of getter contamination, and vacuum pre-stoving at high temperatures, and of cathode temperature were investigated. In all, thirty-two tubes were examined. Table 2 gives the mean spectral emittance values for each batch together with the number of cathodes, and the standard deviation. The variation of emittance with the temperature is shown in Fig. 2. As previously stated, the getter deposit on the bulb caused negligible inhomogeneity in the light distribution. The getter was sufficiently shielded not to cause a measurable increase in absorption of the glass opposite the cathode. Thus there is a real difference in

Table 2. Mean values of  $e_\lambda$  at  $\lambda = 0.66\mu$  for nickel cathodes at varying brightness temperatures

N = number of cathodes		$\sigma$ = standard deviation					
Group	$N$	Room Temperature $e_\lambda$	$\sigma$	$e_\lambda$ at $700^\circ\text{C}$	$e_\lambda$ at $800^\circ\text{C}$	$e_\lambda$ at $900^\circ\text{C}$	$e_\lambda$ at $1000^\circ\text{C}$
H <sub>2</sub> stoved at 900°C; Ba getter ..	20	0.38	0.032	0.39	0.41	0.41	0.45
H <sub>2</sub> stoved at 900°C; Zr getter ..	6	0.34	0.026	0.36	0.34	0.39	0.39
H <sub>2</sub> stoved at 900°C, then vacuum stoved at 1030–80°C for 20 min; Zr getter ..	6	0.41	0.048	0.43	0.42	0.44	0.45

the emittance values between nickel in tubes gettered by evaporated barium, and by non-evaporated zirconium, as indicated by the significance test. Stoving the nickel at high temperatures in vacuum for 20 min also raises its emittance value at room temperature.

The variation of spectral emittance with temperature was similar for all cathodes. The results indicate approximate constancy up to about 800°C brightness temperature, but above 800°C a marked increase with

it was not due to some effect in the reflectometer, a tube was constructed containing a nickel cathode having a platinum/13% rhodium-platinum thermocouple attached. The emittance value was determined by measuring the brightness and thermocouple temperatures simultaneously. The emittance/temperature curve obtained is also plotted in Fig. 2. This cathode had not been polished, but otherwise it was given the same treatment as the hydrogen-stoved batch in zirconium-gettered tubes. Although the emittance was lower the general trend with increasing temperature in the visible range was the same.

**Oxide-coated Cathodes.** Information was obtained on the effect of the barium getter flash on both the measurement of reflectance in general, and on the emittance of the cathode; on the effect of bulb baking temperature on the cathode emittance; and on the variation in emittance for coatings of different surface texture and composition. No variation of emittance with temperature was apparent. Therefore, measurements were made at room temperature in all cases. The mean values for nickel cores, spray coated with barium strontium oxide, are given in Table 1, showing the effects of bulb baking temperature and getter contamination. Table 3 gives some mean values of spectral emittance which have been obtained for different types of oxide-coated cathodes.

If series 1 of Table 1 is examined, it would appear that flashing a barium getter in the tube raises the spectral emittance significantly. If, however, values from series 2 are included when considering cathodes mounted with barium getters, then the difference in emittance between the two types of gettered tubes becomes insignificant. Since the barium getters were mounted in a similar position, the results from these two series were not expected to show a significant difference. The probable explanation is that barium, if deposited in sufficient quantity on the oxide surface raises its emittance, but that in these experiments the effective screening of the cathode from the getter by the mica shield varied from tube to tube, so that varying amounts of barium were deposited on the cathode surface. The wide dispersion of the barium may be brought about by the momentary presence of gas released during the getter flash, this gas being later absorbed by the barium

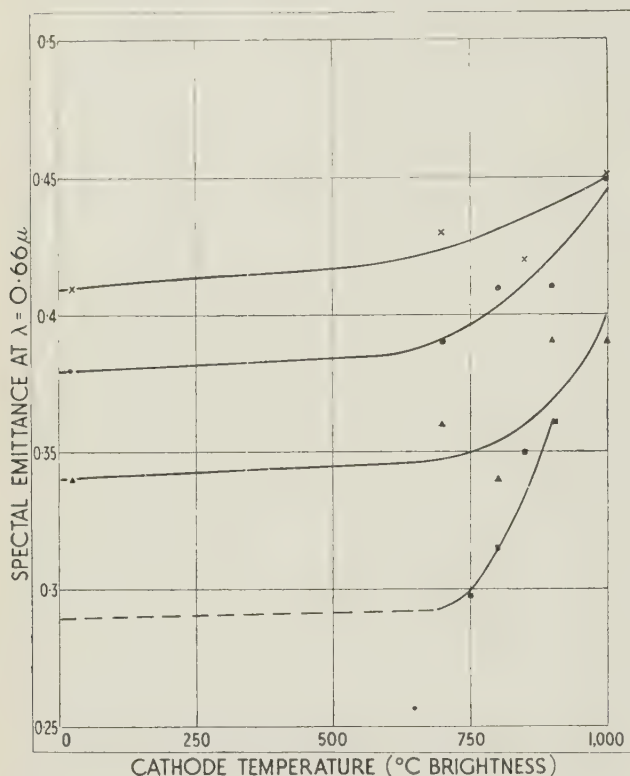


Fig. 2. The variation of spectral emittance with cathode temperature for nickel

- Stoved in hydrogen, barium getter.
- ▲ Stoved in hydrogen, zirconium getter.
- × Stoved in hydrogen, then in vacuum at 1050–1080°C, zirconium getter.
- Unpolished—zirconium getter (thermocouple method).

increasing temperature, giving an average temperature coefficient of  $2.4 \times 10^{-4}$  per deg over the range 800–1000°C. It was not possible to make measurements above 1000°C because of rapid formation of nickel deposits on the glass wall. To confirm the rise in emittance at visible temperatures, and to ensure that

Table 3. Mean emittance values for oxide-coated cathodes of different composition and surface texture. No. of cathodes  $N$ .  $\lambda = 0.66 \mu$

Coating	Getter and baking temperature	$N$	$e_\lambda$
60 $\mu$ rough spray-coated double oxide on nickel tube	Ba or Zr getter		
	Baked at 400°C	7	0.19(4)
	Baked at 300 or 350°C	8	0.14(8)
60 $\mu$ rough spray-coated double oxide on nickel tube	BaMg getter, baked 350 or 400°C	5	0.23(5)
	BaMg getter, unshielded, baked 300°C	5	0.19(2)
	BaMg getter, unshielded, baked 400°C	2	0.44(5)
50 $\mu$ glossy smooth cat. coated double oxide on nickel tube	BaMg getter, unshielded, baked 400°C	3	0.49
50 $\mu$ glossy smooth cat. coated double oxide on nickel tube	BaMg getter, unshielded, baked 300°C	4	0.25(5)
25 $\mu$ glossy smooth cat. coated double oxide on nickel tube	Zr getter, baked 400°C	4	0.15(4)
50 $\mu$ glossy smooth cat. coated triple oxide on nickel	Zr getter, baked 400°C	3	0.10
40 $\mu$ normal cat. coated SrO on Mo strip	Ba getter, baked 400°C	9	0.12
40 $\mu$ normal cat. coated double oxide on Mo strip			
25 $\mu$ normal cat. coated double or triple on 90 $\mu$ W wire			



deposit. This also explains the results for barium-magnesium getter tubes, where a high emittance value, 0.24, was obtained with the unshielded getter whilst a value of 0.13 was obtained when a mica shield was introduced. However, it is interesting to note that the amount of magnesium deposited on the cathode was insufficient to be detected spectrographically.

The effect of the tube baking temperature was first noted in measuring the emittance of smooth cathaphoretic coatings (Table 3). The first tubes constructed were baked on the pumps at 400°C for an hour. The cathodes when formed were grey in appearance with an emittance value of 0.45. Further tubes were baked out at 300°C for two hours, and the cathodes were white with a more normal value of 0.20. This difference was assumed due to some effect of the baking temperature on the coating. A similar effect was found for spray coatings using a nitrocellulose binder, but to a lesser extent. However, there was no significant difference between tubes baked at 300 and 350°C, implying that the effect comes into play at some critical temperature around 400°C. The values in Table 3 refer to coatings of double oxide obtained from the carbonates previously specified and of triple oxide obtained from ammonium precipitated carbonates with a molecular composition of Ba : Sr : Ca = 2 : 1.5 : 1. The general indications are that for equivalent tube-baking temperatures, oxide cathodes prepared from cathaphoretically-coated carbonates have a lower emittance value than those from spray-coated carbonates, while the highest values are obtained with smooth glossy coatings.

#### DISCUSSION

*The Emittance of Nickel.* Numerous values have been quoted for the spectral emittance of nickel, but very few have been obtained on cathode nickel under conditions found in valves. Most of the early published results obtained on nickel in air or inert gases have been summarized by Worthing;<sup>(1)</sup> values from 0.25 to 0.38 are quoted. More recently Weil,<sup>(12)</sup> measuring multi-reflexion from mechanically polished plane nickel mirrors, gave a value of 0.33 at room temperature in fair agreement with Ornstein and Kafoed's<sup>(13)</sup> value of 0.352 measured in an Ulbricht sphere; both measurements were made in vacuum using a copper foil getter. On nickel cathodes, Moore and Allison<sup>(3)</sup> quoted a value of  $e_\lambda = 0.418$  for grade "A" nickel (99% nickel, impurities Co, Fe, Mn, Cu, C) over a temperature range 600–1000°C, whilst Champeix,<sup>(2)</sup> using grade "O" nickel, obtained a value of 0.24 at room temperature, and of 0.44 at 887°C; the getter used was not stated. Johnson and Marshall,<sup>(14)</sup> using a black body method, found an emittance value of 0.41 for so-called clean nickel at 897°C.

The variation of the emittance of nickel with temperature has been investigated by several workers, but the results show considerable disagreement. Theoretically

for low frequencies the spectral emissivity of a metal is given by the expression

$$e_\lambda = 2\sqrt{(c/\lambda\sigma_\lambda)} \quad (8)$$

where  $c$  = velocity of light.

$\lambda$  = wavelength.

$\sigma_\lambda$  = electrical conductivity at wavelength  $\lambda$ .

Since  $\sigma_\lambda$  has a negative temperature coefficient,  $e_\lambda$  will have a positive one.

This relation, however, is known to break down as the visible part of the spectrum is approached. Seitz<sup>(15)</sup> suggested that since the penetration of light into a metal is small, the optical properties in this spectral region would be determined by a thin surface layer of the metal. The temperature coefficient of such a layer would be a function of its nature and thickness. Thus the surface condition of the nickel may well be the main factor controlling the temperature coefficient of emittance. Weil<sup>(12)</sup> found a negative temperature coefficient of  $5.68 \times 10^{-3} \text{ deg}^{-1}$  for  $\lambda = 0.66 \mu$ , whereas Wahlin and Wright,<sup>(16)</sup> Worthing,<sup>(1)</sup> and Moore and Allison<sup>(3)</sup> found no variation of emittance with temperature in the visible region. Price<sup>(17)</sup> obtained a positive temperature coefficient of  $1.7 \times 10^{-4} \text{ deg}^{-1}$  of the same order as the value we obtained above 800°C.

The increase in spectral emittance obtained when the nickel is stoved at a high temperature in vacuum may be due either to a roughening of the surface, or to impurities in the nickel diffusing to the surface and/or evaporating off. In this respect it is significant that the slight contamination with barium from the getter is sufficient to cause an increase of 10–20% in emittance even at 1000°C brightness temperature. From the variation in published results it seems fairly certain that the purity of the nickel and its surface condition have considerable effect on the spectral emittance value obtained, and these factors should therefore be taken into account when using published emittance values.

*Emittance of Oxide-Coated Cathodes.* Few values of spectral emittance have been published for oxide-coated cathodes. Blewett,<sup>(18)</sup> in his review, quoted values of 0.5 to 0.7 for thin coatings without specifying the coating thickness. Moore and Allison<sup>(3)</sup> gave values of 0.287 for spray coatings of 40  $\mu$  thickness on nickel, and 0.392 for a similar coating on Konel. Their measurements were made by a black body method and checked in a reflectometer. Clausing and Ludwig<sup>(19)</sup> obtained values of 0.5 for thin coatings and 0.1 for thick coatings of the order of 150  $\mu$ . This decrease in emittance with increasing coating thickness was substantiated by Champeix<sup>(2)</sup> with both spray and cathaphoretically coated cathodes. He found an emittance of 0.27 at 30  $\mu$  decreasing to 0.09 at 105  $\mu$  for spray coatings, and 0.31 at 15  $\mu$  decreasing to 0.18 at 80  $\mu$  for cathaphoretic coatings. For 50  $\mu$  double oxide coatings formed from carbonates sprayed on nickel, Herrmann<sup>(20)</sup> obtained values of 0.18–0.30 using a spot of known emissivity,

while Liebold<sup>(21)</sup> found the value 0.25 over the range 777–977°C by pyrometric and retarding field current method. In view of the range of values found in our present investigation (Tables 1 and 3) very little comparison with results of previous workers is possible, since their conditions were incompletely specified. Furthermore, it is uncertain whether corrections were made for transmission of the glass bulbs. However, the value obtained by Champeix,<sup>(2)</sup>  $e_\lambda = 0.20$  for spray coatings 65  $\mu$  thick is in fair agreement with our results in barium gettered tubes of  $e_\lambda = 0.17(5)$  for similar coatings of thickness 60  $\mu$ .

#### CONCLUSIONS

The average value of the spectral emittance for electrolytically polished "O" grade nickel which had been stoved in hydrogen and mounted in zirconium gettered tubes was 0.34 at room temperature at  $\lambda = 0.66 \mu$ , with a standard deviation of 0.03. Flashing a barium getter raised this average value to 0.38, standard deviation 0.03. Vacuum stoving the nickel at a high temperature, 1050°C for 20 min, also raised the emittance value, giving 0.41 with standard deviation 0.05. The variation of the emittance with temperature had the same general characteristics in all cases, namely, constant emittance from room temperature to 800°C and then a marked rise to 1000°C brightness temperature with an average temperature coefficient of  $2.4 \times 10^{-4} \text{ deg}^{-1}$ .

For oxide-coated cathodes prepared from equimolar barium strontium carbonates sprayed to a thickness of 60  $\mu$  on to "O" grade nickel the emittance value was found to depend on the baking temperature and getter used. For tubes baked below 400°C and gettered with a non-evaporating zirconium wire, the average value was 0.15, standard deviation 0.04.

Flashing a barium getter tended to increase this value if the getter was not well screened, and in the case of unshielded magnesium-barium getters a value of 0.24 was found. The actual deposit on the glass wall does not affect the measurement of reflectance in a reflectometer. Baking the tubes at 400°C raised the average emittance values by 0.04. The coating composition, texture, and method of coating also affects the emittance value markedly. Values ranging from 0.1 to 0.5 were obtained under different conditions.

#### ACKNOWLEDGMENTS

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# Studies of the viscosity and sedimentation of suspensions

## Part 2.—The viscosity and sedimentation of suspensions of rough powders

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[Paper first received 26 June, 1950, and in final form 14 September, 1950]

The viscosity of suspensions of rough methyl-methacrylate powders in aqueous solutions is shown to increase at all concentrations as the particle size is reduced. This is explained on the assumption that the individual particles behave as if surrounded by a layer of immobile liquid, held in the irregularities of their surfaces. The thickness of the layer, for a particular material, is independent of the viscosity of the suspending liquid, the concentration of the suspension and the size of the particles above a certain minimum value below which it decreases continuously with particle size. Sedimentation experiments made on suspensions of the same particles confirm these conclusions.

In an earlier paper<sup>(1)</sup> it was shown that the relative viscosity of a suspension of smooth spheres is independent of their absolute size but dependent, to some extent, upon their size distribution. For other shapes of particles and for roughly-surfaced materials, however, these conclusions do not apply. Oden<sup>(2)</sup> found an increase in the relative viscosity of sulphur sols with decreasing size at a given volume concentration. Pryce-Jones,<sup>(3)</sup> De Vaney and Shelton<sup>(4)</sup> and Ward (H) and Kammermeyer<sup>(5)</sup> all report similar effects at different size ranges with a variety of materials; thus, Oden worked with particles of millimicron size, Pryce-Jones at about 5 microns and De Vaney and Shelton at about 50 microns, none using spherical particles. Hatschek<sup>(6)</sup> suggested an explanation of Oden's results by postulating adsorption layers on the particles but no explanation of the results of the other workers has been offered. The theoretical work of Jeffery<sup>(7)</sup>, McBain,<sup>(8)</sup> Burghers<sup>(9)</sup> and Kuhn<sup>(10)</sup> shows that, for particles at a given volume concentration and not exhibiting Brownian motion, the relative vis-

cosity increases with the asymmetry of the particles. Increasing particle asymmetry with decreasing particle size might, therefore, explain the observed results. Alternatively, adsorption or trapping of suspending liquid by the particles (leading to an apparent increase in the volume of the solid phase) or the angularity of the particles (as suggested by Steinour<sup>(11)</sup>) might be important. These possibilities were explored in the series of experiments described below.

### THE SUSPENSIONS

These were identical with those used in the earlier experiments<sup>(1)</sup> except that the methyl-methacrylate polymer spheres were replaced by a rough-surfaced, irregularly-shaped form of the plastic. Six samples, details of which are given in Table 1, were prepared from the raw powder. Photo-micrographs of Samples (a) and (e), taken with black-background illumination, are shown in Figs. 1 and 2. It was assumed in the subsequent experiments that their surface-chemical properties were the

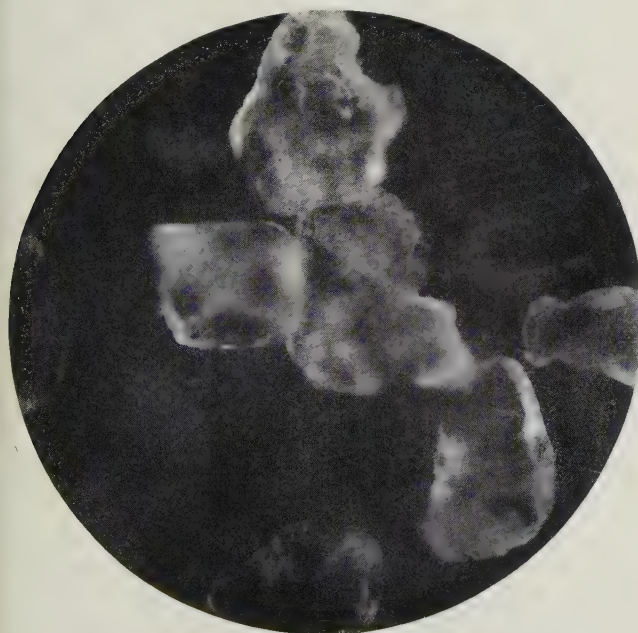


Fig. 1. Photomicrographs of powder, sample (a)

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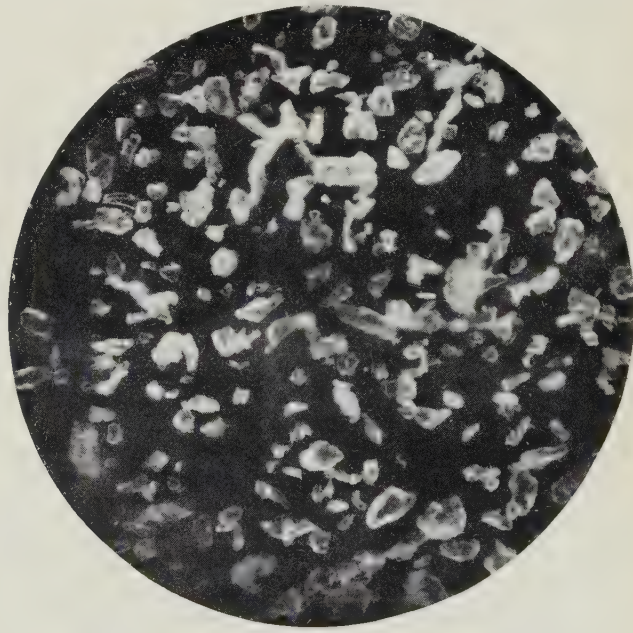


Fig. 2. Photomicrographs of powder, sample (e)

same as for the spherical particles and any differences in their behaviour when in suspension could be attributed solely to additional physical effects. The suspending liquid—an aqueous solution of lead nitrate and glycerol—had a viscosity of about 6 centipoises and a density equal to that of the suspended particles.

#### MEASUREMENT OF VISCOSITY

The rising-sphere viscometer described in earlier experiments<sup>(1)</sup> was used, four spheres of about 1.4 mm diameter and made from acrylic and styrene plastic mixtures, pigmented for identification, being employed. The apparatus was immersed in a thermostat maintained at  $25^{\circ}\text{C} \pm 0.05^{\circ}$ . The volume concentration of each sample was increased progressively from zero to about 25% and at each concentration the relative viscosity was measured by releasing spheres from two different depths and timing their rise to the suspension surface. All air bubbles were first removed from the suspensions by subjecting them to a vacuum.

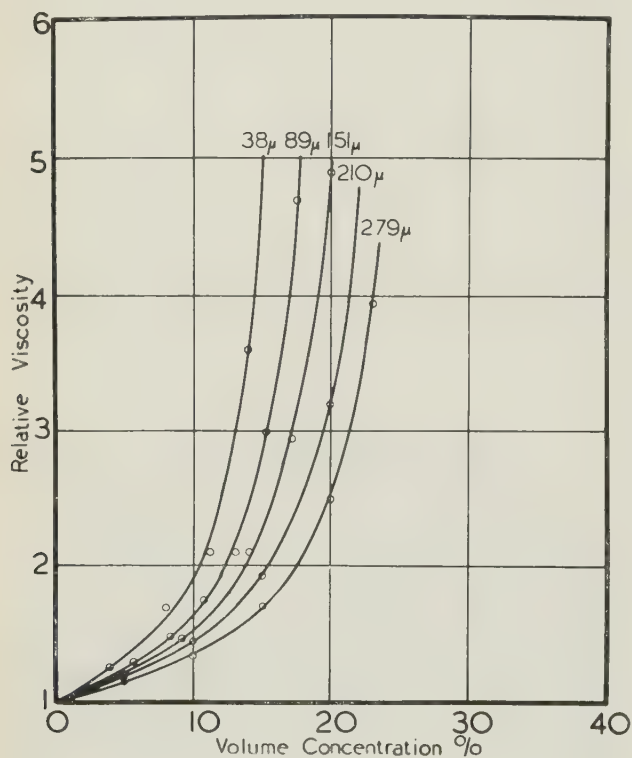


Fig. 3. Relationship between relative viscosity and volume concentration of methacrylate powders of various mean statistical diameters

**Results.**—The rising velocity of each sphere at each volume concentration was calculated as before,<sup>(1)</sup> after applying the Faxen correction for the wall effects. The highest Reynolds number attained by any sphere was 1.6, but in the majority of cases a value of 0.5 was not exceeded. It is estimated that the resulting measure-

ments were accurate to within  $\pm 2\frac{1}{2}\%$ . Fig. 3 shows the variation of relative viscosity with volume concentration for the five powders (a) to (e) inclusive.

**Discussion.**—The results confirm the experiments of Pryce-Jones,<sup>(3)</sup> De Vaney and Shelton<sup>(4)</sup> and Ward (H) and Kammermeyer<sup>(5)</sup> in that, in contrast to suspensions of spheres, the relative viscosity of a suspension of rough, irregularly-shaped particles at a given concentration is dependent upon the size of the particles, increasing with decreasing average-particle size. This variation cannot be explained entirely by the asymmetry of the particles; Burghers' theoretical results<sup>(9)</sup> would require the axial ratios to vary from about 5 : 1 for the coarsest powder to 35 : 1 for the finest, which Table 1 shows they did not.

Table 1. Details of suspension powders

Sample	B.S.S. sieve size	Mean statistical diameter $\mu$	Ratio Length Breadth (average)	Specific surface ( $\text{cm}^2$ )
(a)	72-85	279	1.6	302
(b)	100-120	210	2.0	518
(c)	150-170	151	2.2	824
(d)	200-300	89	2.3	1 494
(e)	-300	38	2.3	3 547
(f)	72-170	—	—	497

Specific-surface measurements were made by a microscopic method developed by Needham and Hill.<sup>(13)</sup> It has the advantage of giving the envelope area of the particles.

do. The slopes of the relative viscosity/volume concentration curves at infinite dilution ( $k$  in Einstein's equation) vary as the square-root of the surface area and on extrapolation to zero surface-area, i.e. when surface effects are contributing nothing to the forces opposing shear, a value of  $k$  of 1.85 is obtained. This is almost identical with the value of  $k$  obtained for wide size-range spheres.<sup>(1)</sup> Fig. 4 shows that the relative viscosity at any concentration varies practically linearly with the specific surface of the powder. When the relative viscosities of the wide size-range sample (f) were plotted on the graph they lay on the lines already drawn for the narrow size-range samples (a) to (e) inclusive. Thus, again in contrast to suspensions of spheres, the size distribution of a rough powder has little effect upon its relative viscosity in suspension. Microscopic examination shows, however, that a sample of rough particles, closely sized by sieving, will cover a much wider range than a similarly sized spherical powder when considered on a particle-volume basis. When the curves are extrapolated back to zero specific surface, i.e. surface effects are eliminated, the values of the viscosity of a suspension of wide size-range spheres are obtained.

The assumption was then made that the viscosity of the suspension arose from: (1) The presence of the particles in the liquid, assumed equal to that for wide size-range spheres. (2) Immobile liquid held in the irregularities of the particle surfaces increasing the



effective volume-concentration in suspension. No allowance for the asymmetry of the particles was made because its effect would not be large for the axial ratios of about 2:1 employed. The average thicknesses of immobile liquid held to the surfaces of the particles of each sample at various concentrations were calculated and are recorded in Table 2.

Table 2. Thickness of stagnant liquid layer on rough particles

Sample	Mean statistical diameter ( $d$ ) $\mu$	Volume concentration per cent					Mean stagnant layer thickness ( $L$ ) $\mu$	Ratio $\frac{d}{L}$
		5	10	15	18	20		
(a)	279	6.71	6.15	5.78	6.37	6.98	6.4	43.7
(b)	210	7.17	6.68	6.18	6.52	6.92	6.7	31.3
(c)	151	6.77	6.05	5.73	6.43	6.93	6.4	23.6
(d)	89	5.08	4.35	4.29	5.09	—	4.7	18.8
(e)	38	3.33	2.47	2.91	—	—	2.9	13.1

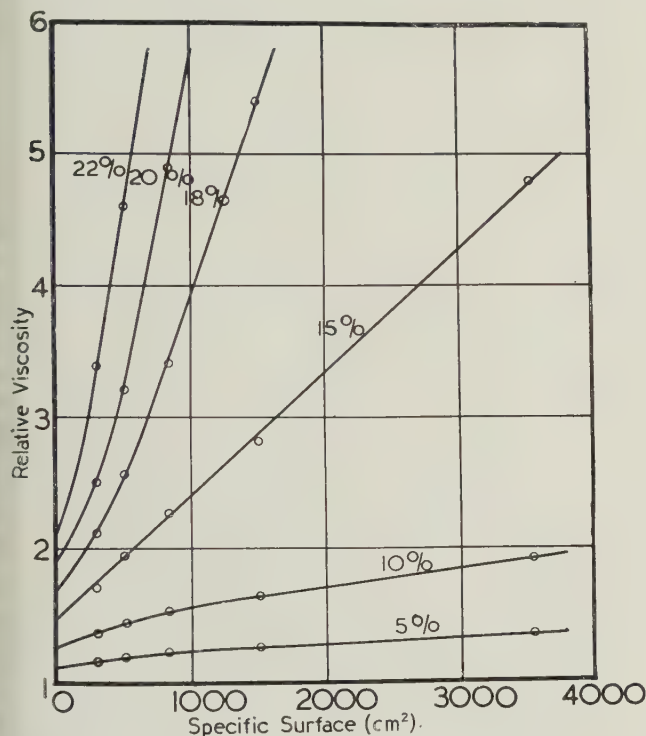


Fig. 4. Variation of relative viscosity with specific surface of methacrylate powders at various volume concentrations

The following properties can be attributed to the stagnant layer: (1) For a particular powder it remains of almost uniform thickness over a wide range of volume concentrations. (2) For coarse samples the layer is of practically the same thickness, but for smaller particles the thickness decreases with decreasing particle size.

This suggests that the maximum thickness of the layer is determined by the degree of roughness at a given point on the surface of the particular material. As the particles are reduced in size, the effect of their edges, where stagnant liquid will be continually swept away, becomes

increasingly important and the average layer thickness is reduced. The thickness, relative to the particle diameter, however, increases continuously with decreasing particle size.

#### SEDIMENTATION EXPERIMENTS

If the density of the suspending liquid is reduced and the particles allowed to sediment, it would be expected that the immobile-liquid layer would still be present and could be detected by measurements made on the settling rate of the suspension. The sedimentation liquid consisted of an aqueous solution of lead nitrate and glycerol as used in the viscosity measurements but the density was reduced to 1.10 g/ml and the viscosity to about 3.5 centipoises. The tests, which were similar to those described by Steinour,<sup>(11)</sup> were made in straight-walled, flat-bottomed tubes 180 mm tall and 30 mm internal diameter immersed in a thermostat maintained at  $25^{\circ}\text{C} \pm 0.05^{\circ}$ . All air bubbles were removed from the suspension by reducing the air pressure and, after thoroughly stirring, readings of the height of the suspension were taken at regular intervals with a micrometer microscope. Plots of fall of the upper boundary against time were made and found to be linear in all cases, the constant of proportionality giving the settling rate of the suspension at the given volume concentration. Measurements were made at concentrations from 10 to 35% by volume, in steps of 5%. The results were evaluated by a method first used by Steinour.<sup>(11)</sup> He showed that if  $w_i$  = volume of immobile liquid/unit volume of hydrodynamic volume-concentration,  $c$  = volume concentration of suspended particles,  $Q$  = settling rate at volume concentration  $c$ , then

$$1 - \frac{c}{1 - w_i} \propto \log_{10} Q \left( \frac{1 - w_i}{1 - c - w_i} \right)^2$$

where the constant of proportionality,  $p = 1.82$ .

Needham and Hill<sup>(12)</sup> have found the theory unsatisfactory but, since it is derived for the viscous region of flow and they used it at Reynolds numbers up to 50, the disparities which they found are not surprising.

**Results.**—From the experimentally determined settling rates of the four powders, plots of  $1 - \frac{c}{1 - w_i}$  against

$\log_{10} Q \left( \frac{1 - w_i}{1 - c - w_i} \right)^2$  (Fig. 5) were made, suitable values of  $w_i$  being found by trial and error. In no case, however, did the points lie exactly on a straight line and an estimation of the average slope of 1.82 was necessary. According to Steinour, the value of  $w_i$  should have been the same for each sample but this was not found to be so; instead it increased with decreasing particle size. From a knowledge of the specific surface of each powder, the average thickness of immobile liquid on each of the powders was calculated and is compared with that obtained from viscosity measurements in Table 3.

Sedimentation tests made in aqueous lead nitrate solution of viscosity about 1 centipoise gave identical  $w_i$  values with those obtained at 3.5 centipoises and, since the viscosity measurements were made at 6 centipoises, it is apparent that the thickness of the layer of immobile liquid is substantially independent of the suspension-liquid viscosity. The layers are more than one hundred times thicker than would be expected on the assumption of adsorption at the surface and as no such effects were found with spherical particles of the same material,<sup>(1)</sup> Steinour's statement that immobile liquid is not held to the surface by adsorption is confirmed. His conclusion that the amount of immobile liquid is dependent upon particle shape but independent of its size is not, however, borne out. As his experiments were made on three emeries all from different sources, it is probable

that the shape and surface characteristics were different in each case.

### CONCLUSIONS

The increase in viscosity of suspensions of rough powders is mainly due to liquid held in the irregularities of their surfaces which increases their effective volume. There is no evidence to suggest that the liquid is adsorbed and the layers can be so thick that any theories of chemical binding of the liquid to the particles become untenable. The thickness of the layer is dependent upon the degree of surface roughness but not on the viscosity of the liquid. For a particular material, the thickness of the layer is substantially constant above a certain particle size below which it decreases continuously although the ratio of thickness of surface layer to size of particle increases continuously with decreasing size. The size distribution of the particles has an inappreciable effect upon the viscosity of the suspension.

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(Part 1 of this paper, "The viscosity of suspension of spherical particles," appeared in the November issue of this Journal.

Table 3. Thickness of stagnant layers on rough powders

Sample	Method of measurement	
	Sedimentation $\mu$	Viscosity $\mu$
(a)	6.98	6.4
(b)	6.68	6.7
(c)	6.35	6.4
(d)	4.72	4.7

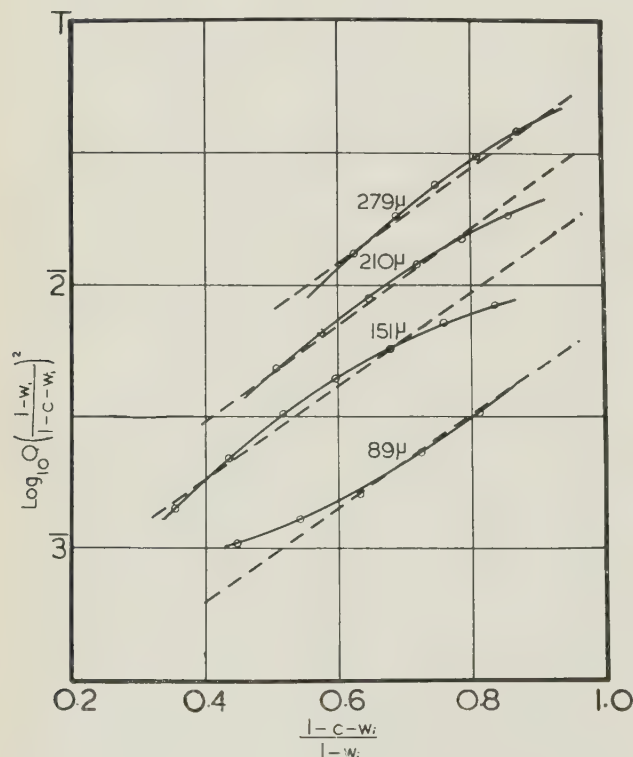


Fig. 5. Relationships for methacrylate powders of various mean statistical diameters



# Errors in diffusion measurements by the Loschmidt method

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[Paper received 14 August, 1950]

The problem of obtaining diffusion coefficients of gases with high absolute accuracy is considered. It is shown that the Loschmidt method is capable of giving such results. The diffusion coefficient is obtained from the formula:—

$$D = \frac{\pi L^2}{4(t_1 + 0.424t_2 + t')} \left[ \frac{1}{(f+1)^2} \pm \frac{2}{r(f+1)} \right]$$

where  $L$  is the total length of the tube,  $f$  the ratio of concentrations of the reference substance in the two half-tubes and  $t_1$  the time of diffusion.  $t_2$ ,  $t'$  and  $r$  are correction factors. The conditions which are likely to yield the highest accuracy are established.

The work of Hirschfelder and others<sup>(1,2)</sup> has recently directed attention to the problem of determining diffusion coefficients in the gaseous phase with high absolute accuracy. Following these authors it is now possible to calculate from diffusion coefficients the two constants  $\epsilon$  and  $r_0$  in the Lennard-Jones potential between spherical non-polar molecules. This potential is of the well-known form:—

$$E(r) = 4\epsilon[(r_0/r)^{12} - (r_0/r)^6]$$

The constants have hitherto been determined from analysis of second virial coefficient data and more recently from viscosity measurements.<sup>(2)</sup> As the viscosity is not particularly sensitive to the form of the assumed law of force between the molecules, ordinary diffusion has recently been suggested<sup>(2)</sup> as a suitable alternative source of information about the magnitude of  $\epsilon$  and  $r_0$ . It is possible to show by reference to the functions tabulated by Hirschfelder *et al.* that if the diffusion coefficient is known to within a few parts in a thousand, the pressure is measured with  $\sigma_p = 0.1$  mm of mercury and the temperature with  $\sigma_T = 0.01^\circ \text{C}$ ,  $\epsilon$  and  $r_0$  could be obtained with an absolute fractional accuracy of about  $\pm 1\%$  and  $\pm 0.5\%$  respectively. The corresponding accuracies of the constants obtained from virial coefficient data are somewhat worse and no virial coefficient data is available for mixed gases, whereas diffusion offers a particularly straightforward means of studying the law of force between unlike molecules.

The determination of self-diffusion coefficients in non-ideal gases, work which is in progress in this laboratory, is of interest from the point of view of kinetic theory. As the deviations from ideal conditions are second-order effects, the diffusion coefficients have to be determined with high absolute accuracy to make interpretation of the results possible.

## THE LOSCHMIDT METHOD

A relatively simple case of one-dimensional diffusion of two substances occurs under the following boundary conditions. The diffusion chamber is a linear tube of uniform cross-section and of length  $L$ . The concentration of the reference substance at time  $t = 0$  is  $c_0$  between

$0 \leq x \leq h$ . At time  $t$  is the concentration  $c$  at the point  $x$  ( $0 \leq x \leq L$ ) will be given by

$$c = c_0 \left[ (h/L) + (2/\pi) \sum_{n=1}^{\infty} n^{-1} \exp(-m^2 Dt) \cos(mx) \sin(mh) \right] \quad (1)$$

with  $m = n\pi/L$ . If  $h$  is so chosen that it is a definite integer fraction of  $L$ , e.g.  $(L/h) = s^{-1}$ , then the equation is easily solved and it is possible to calculate how much of the reference substance at time  $t$  is to be found in each layer of thickness  $h$ . The case  $s = 1/2$  forms the basis of the "Loschmidt method" of determining diffusion coefficients and is generally recognized as more suitable for accurate work in the gaseous phase than any other experimental procedure which has been proposed. After diffusion is terminated by separating the half-tubes the contents are analysed and the results give the average concentrations of the reference substance in each chamber. Hence, for convenience, (1) has to be cast in a different form.

Let the average concentration of the reference substance after time  $t$  be  $u$  between  $0 \leq x \leq h$ , then

$$u = h^{-1} \int_0^h c dx = c_0 \left[ (h/L) + (2L/\pi^2 h) \sum_{n=1}^{\infty} n^{-2} \exp(-m^2 Dt) \sin^2(mh) \right] \quad (2)$$

For the case of  $s = 1/2$  and putting  $c_0 = u + o$  after some expansion

$$(u - o)/(u + o) = 1 - \psi(v) = 8\pi^{-2} [e^{-v} + (1/9)e^{-9v} + (1/25)e^{-25v} + \dots] \quad (3)$$

where  $v = \pi^2 Dt/L^2$ . Tables of the function  $\psi(v)$  were given by McKay<sup>(3)</sup> to a mechanical accuracy of one part in a thousand. It can also be shown<sup>(3)</sup> that to a good approximation

$$\psi(v) = 4\sqrt{(v/\pi^3)} \quad (4)$$

provided that  $v$  is sufficiently small. For  $v = 0.5$  the approximation is to within 3 in 5000 and for lower values of  $v$  the error is further diminished. Clearly, this simple formula is useful in diffusion work with gases.

For a case where  $L = 10^2$  cm,  $t$  may vary between 100 and 4 000 sec, say, provided the diffusion coefficient varies between 0.1 and 0.01 cm<sup>2</sup>/sec. Thus  $10^{-3} \leq v \leq 10^{-1}$  and (4) is always obeyed. We obtain

$$D = \pi L^2 / 4t(f+1)^2 = (\pi L^2 / 16t)(y-1)^2 \quad (5)$$

where

$$f = u/o, \quad y = (u-o)/(u+o)$$

In the ensuing discussion we shall use (5) as (3) is totally unsuitable for analysis. It will also be assumed that the gas under consideration is perfect. As this assumption only enters in the calculation of correcting factors which are themselves small it will be permissible. In order to quote numerical results for the errors which arise we shall tentatively assume  $L = 10^2$  cm,  $D = 0.1$  cm<sup>2</sup>/sec and  $t = 10^3$  sec so that  $f = 7.87$ .

#### CORRECTION TERMS

The equation (3) refers to an apparatus with  $s = 1/2$ . In practice this condition will never be exactly satisfied partly because the two tubes would rarely be identical in length and partly because valve attachments to one or both of the tubes may add capillary sections to the original length of the tubes. The effective contribution of capillary sections to the length of a tube is best assessed by weighting the length of the capillary with the cross-section ratio. To establish the correction factor arising from a slight deviation from  $s = 1/2$ , let the value of  $h/L$  in (2) be

$$h/L = 1/2 \pm 1/r \quad (6)$$

where  $r$  is a large integer such that  $1/r$  approximates most closely to the measured correction to  $s = 1/2$ . Then from (2) and (6)

$$y = \pm 2/r + 8\pi^{-2}[e^{-v} + (1/9)e^{-9v} + \dots] \quad (7)$$

where terms in  $(1/r)^2$  and higher are neglected and the approximation formula  $\sin^2(A \pm x) = \sin^2 A \pm 2x \sin A \cos A + x^2 \cos^2 A$  is used which is valid for  $x$  small compared to unity. Combining (7) with (4) and (5) and again neglecting terms in  $(1/r^2)$  we have finally

$$D = (\pi L^2 / 4t) \left[ \frac{1}{(f+1)^2} \pm \frac{2}{r(f+1)} \right] \quad (8)$$

Let  $h = 50.1$  cm and  $L = 100$  cm then the deviation from  $s = 1/2$  is 1 in 1 000 and  $r = 10^3$ . Equation (8) shows that neglect of the term in  $r$  can cause a constant error of 2% in the value of the coefficient of diffusion.

An error is introduced by the existence of a small initial pressure difference between the contents of the two halves of the diffusion chamber. Let the two halves be labelled 1 and 2 respectively, then, since the volumes are assumed equal,  $p_1 = n_1 RT/V$  and  $p_2 = n_2 RT/V$ . Shortly after the tubes are brought in contact some gas is transferred by "blow-over" to the low pressure side

until a uniform pressure  $p$  is established throughout the diffusion chamber. Let  $p_1 > p_2$  and  $p_1 - p_2 = \Delta$ , then  $p_1 - p = \Delta/2 = p - p_2$ . Clearly, the amount of gas transferred to side 2 is  $(n_2 - n_1)/2 = (V/RT)\Delta/2$ .

A case of interest occurs when the high pressure side contains both gases  $a$  and  $b$ , the former being the reference substance, while the low pressure side contains only  $b$ . Prior to contact between the tubes the molar fraction of the reference gas at the high pressure side will be  $x_{1a}$ . It will be shown that the "blow-over" takes place in an exceedingly short time, while contact is established between the tubes, during which no appreciable diffusion occurs. Let contact be broken between the tubes as soon as the pressure difference is equalized. Then

$$y = (x_{1a} - x_{2a})/(x_{1a} + x_{2a}) = (1 + \Delta V/n_{2b}RT)^{-1} \quad (9)$$

and combining with (5)

$$t' = (\pi L^2 / 16D)\Delta^2/p^2 \quad \text{since } V/n_{2b}RT = p - \Delta/2 \quad (10)$$

The speed of the "blow-over" is sufficiently great for  $t'$  to represent a correction factor to the measured time  $t$  in a diffusion run. This error will be most important at low pressures and from (10) one can obtain an idea of the magnitude of  $\Delta$  which is permissible. Using  $p = 5$  cm of mercury and  $\Delta = 0.1$  mm of mercury  $t' = 0.1$  sec, a figure which is less than a likely standard deviation for  $t$ . However if  $\Delta = 1$  mm of mercury for the same pressure the error  $t' = 10$  sec.

Hydrodynamic theory predicts the rate of "blow-over"

$$m = A\sqrt{(\Delta\rho)} \quad (11)$$

where  $m$  is the flux in g/sec,  $A$  the cross-sectional area of the tubes and  $\rho$  the density of the gas on the high pressure side in g/c.c. Clearly, at any time  $t$ ,  $m_1 = A(\Delta_1\rho)^{1/2}$ . Now

$$\begin{aligned} p_1(t+dt) &= p_1(t) - ARTV^{-1}\sqrt{(\Delta_1\rho)}dt \\ p_2(t+dt) &= p_2(t) + ARTV^{-1}\sqrt{(\Delta_1\rho)}dt \end{aligned}$$

$$\text{i.e.} \quad d\Delta_1/dt = -2ARTV^{-1}\sqrt{(\Delta_1\rho)} \quad (12)$$

integrating with  $\Delta_1 = \Delta$  at  $t = 0$  and setting  $\Delta_1 = 0$  at  $t = t_\infty$  we have:

$$t_\infty = (V/ART)\sqrt{(\Delta/\rho)}$$

$$\text{and } \sqrt{\Delta_1} = \sqrt{\Delta} - (ART/V)\sqrt{(\Delta/\rho)} \quad (13)$$

Since the average flux  $\bar{m}$  can be seen to be  $(A/2)\sqrt{(\Delta\rho)}$  the total amount of gas transferred to the low pressure side is

$$\bar{m}t_\infty = (V/RT)\Delta/2 \quad (14)$$

in agreement with the previous result. Using (13) with  $V = 150$  c.c.,  $p = 5$  cm of mercury and  $\Delta = 0.1$  mm of mercury we verify that the "blow-over" is of exceedingly short duration,  $t_\infty = 5A^{-1} \cdot 10^{-3}$  sec at room temperature and decreases with the inverse square root of the



pressure. The effect will, therefore, be over well before the two halves of the tubes are fully in alinement. The "blow-over" will tend to produce a small amount of mixing near the entrance of the low pressure side as a result of the unsymmetrical injection of a stream of gas from the high pressure side rather than a uniform displacement of the boundary line between the contents of the two tubes towards the low pressure side. Consequently, for small  $\Delta$ , such as is envisaged, a type of correction as in (10) would be permissible, since the amount of mixing caused by the hydrodynamic effect would not materially alter the final value of  $D$ . The error caused by "blow-over" is thus reduced to an error in the time and it is clear that even in an unfavourable case ( $p = 5$  cm of mercury) a pressure difference of 0.1 mm of mercury is permissible.

For a short period at the beginning and at the end of each diffusion measurement the two halves of the diffusion chamber are in incomplete alinement. It will be shown that depending on the speed with which the two tubes are alined, neglect of the consideration of partial overlap may give rise to a constant error of 1 to 2 sec in the value of  $t$ .

The cross-section of the tubes is circular and a position of partial overlap such as occurs twice in every run, may be described by specifying the radius  $r$  of the cross-section and the distance  $\delta$  between the centres of the circles. Simple trigonometric considerations give the area of overlap  $A_0$  as

$$A_0 = 2r^2 \cos^{-1}(\delta/2r) - \frac{1}{2}\delta\sqrt{(2r^2 - \delta^2)} \quad (15)$$

Since an alinement involves the variation of  $\delta$  between 0 and  $2r$  the average value of the common area,  $\bar{A}_0$ , is

$$\bar{A}_0 = 4r^2/3 \quad (16)$$

The averaging implicitly assumes that all values of  $\delta$  are equally weighted, i.e. that the two tubes are brought into alinement with a constant velocity. This is a practical approximation not likely to be far from the real state of affairs. Let the time during which there is a total alinement be  $t_1$  and the time during which there is partial overlap be  $t_2$ . Clearly, in order to obtain the total time  $t$  each of these times must be weighted with the area of contact to which they refer. Thus

$$\pi r^2 t = \pi r^2 t_1 + (4r^2/3)t_2 \quad \text{or} \quad t = t_1 + 0.424t_2 \quad (17)$$

The sum of the times of partial alinement,  $t_2$ , will, in practice, be 2 to 4 sec.

#### OPTIMUM CONDITIONS

It is possible to calculate the ratio  $(1+f)/t$  which will give the maximum accuracy in  $D$  for given standard deviations  $\sigma_f$  and  $\sigma_t$  which are assumed to be known. Equation (5) can be written

$$x = (D/L^2) = (\pi/4t)(f+1)^{-2} \quad (18)$$

and it will be observed that the left-hand side of this equation is constant in any given run. Furthermore,

$$\sigma_x^2/x^2 = F_x^2 = x^{-2}(\partial x/\partial t)^2\sigma_t^2 + x^{-2}(\partial x/\partial f)^2\sigma_f^2 \quad (19)$$

To obtain a stationary value of  $F_x^2$ , say,  $F_{xm}^2$ , subject to the subsidiary condition (18), we put

$$\left. \begin{aligned} \partial F_{xm}^2/\partial t + K\partial\phi/\partial t &= 0 \\ &= -2t_m^{-3}\sigma_t^2 - K\pi/4t_m^2(f_m+1)^2 \\ \partial F_{xm}^2/\partial f + K\partial\phi/\partial f &= 0 \\ &= -8\sigma_f^2/(f_m+1)^3 - 2K\pi/4t_m(f_m+1)^3 \end{aligned} \right\} \quad (20)$$

where  $\phi = x - (\pi/4t)(1+f)^{-2}$  and  $K$  is the Lagrangian undetermined multiplier which can be eliminated with the aid of the set (20) leading to

$$\sigma_f^2/\sigma_t^2 = (f_m+1)^2/2t_m^2 \quad (21)$$

The equation (21) in conjunction with (18) leads to definite values of  $t$  and  $f$ , namely,  $t_m$  and  $f_m$  which are the best components provided it can be shown that  $F_{xm}^2$  represents a minimum. In order to do that let us seek an  $(F'_x)^2$  in the immediate neighbourhood of  $F_{xm}^2$  by setting

$$x = \frac{\pi}{4(t_m + \alpha)(f_m - \beta + 1)^2} \quad (22)$$

where  $\alpha$  and  $\beta$  are very small. By expanding (22) it is easily seen that neglecting second and higher powers of the small quantities

$$\alpha = 2t_m\beta(f_m+1)^{-1} \quad (23)$$

Introducing the factors  $\alpha$  and  $\beta$  in (19) to form  $(F'_x)^2$  and expanding as far as the second powers of small quantities it is seen that

$$\begin{aligned} (F'_x)^2 = F_{xm}^2 - & \left[ \frac{2\alpha}{t_m^3}\sigma_t^2 - \frac{8\beta}{(f_m+1)^2}\sigma_f^2 \right] \\ & + \left[ \frac{3\alpha^2}{t_m^4}\sigma_t^2 + \frac{12\beta^2}{(f_m+1)^4}\sigma_f^2 \right] - \dots \quad (24) \end{aligned}$$

Using (23) and (21) the first term in the small quantities disappears and

$$(F'_x)^2 = F_{xm}^2 + \left[ \frac{12\beta^2}{t_m^2(f_m+1)^2}\sigma_f^2 + \frac{12\beta^2}{(f_m+1)^4}\sigma_f^2 \right] - \dots \quad (25)$$

Clearly, an arbitrarily small variation in the values of  $t_m$  and  $f_m$  will give a value of  $F_x^2$  which is in excess of  $F_{xm}^2$ , hence the condition (21) represents a minimum. With  $\sigma_f = 0.01$  and  $\sigma_t = 0.5$  sec the optimum set is  $t_m = 415$  sec and  $f_m = 12.8$ , the fractional accuracy of  $f$  being 0.08%.

The foregoing analysis applies to the case when the standard deviation of  $f$  is independent of the value of  $f$ . This would be the case in most instances of ordinary chemical or physical analyses of the sample or with radioactive measurements provided they are carried out with integrating instruments. If a Geiger tube is used,

the foregoing results do not apply since in that case the errors in  $f$  depend on  $f$ .

Taking account of the various correction terms the final formula to be used with the Loschmidt method for the calculation of the coefficient of diffusion is

$$D = \frac{\pi L^2}{4(t_1 + 0.424t_2 + t')} \left[ \frac{1}{(f+1)^2} \pm \frac{2}{r(f+1)} \right] \quad (26)$$

Using the principle of equal effects it is possible to obtain from (26) in conjunction with  $D = 0.1$  and the values of  $t_m$  and  $f_m$  obtained above a set of standard deviations for the various quantities involved in (26). These values should be aimed at in order to attain a given accuracy in  $D$ . For a standard deviation of, say, 3 parts in 1 000 in  $D$  it is easily shown that the errors likely to arise in the measurement of  $r$  and  $t'$  have a negligible effect. The set obtained is

Table 1

	$L$ (mm)		$t_1$ (sec)	$t_2$ (sec)
$\sigma$	$\pm 0.56$	$\pm 0.011$	$\pm 0.36$	$\pm 0.30$

so that  $\sigma_t = 0.47$  sec. With the exception of  $\sigma_f$ , all these standards deviations are easily attainable in practice. This stresses the need for an accurate method of analysis.

In the foregoing discussion the effect of vibrations

and the effect of a temperature gradient have not been treated. The quantitative assessment of the influence of vibration on the diffusion process is not feasible. The practical elimination of vibrations of high frequency should be possible by using a bulky apparatus and floating it in some viscous thermostating fluid. A temperature gradient along the longitudinal axis of the apparatus may be responsible for an initial pressure difference between the two halves of the diffusion chamber and this effect has been dealt with. It would be possible to investigate the influence of convection on the diffusion process. Convection would arise if, for a vertical position of the diffusion tube, the top part of the apparatus were colder than the bottom part. This situation is not, however, likely to arise in practice.

#### ACKNOWLEDGMENT

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## Correspondence

### Survey of General and Applied Rheology

Dr. Harrison objects to certain aspects of Dr. Lawrence's review of Dr. Scott Blair's book on rheology, and the words "occult," "mystical" and "esoteric" fly freely about in your columns.

In the natural conduct of physics, one experiments and endeavours to explain the results obtained by developing an argument or theory leading to the logical expectation of the data. Failing sufficient knowledge or mental capacity at the present state of evolution of the human race to develop such a logical theory in a most complicated and difficult domain (of which no one need be ashamed), there arises the possibility of resort to empiricism, using any arbitrary mathematical formula. One such is the so-called power law of the form  $x = yt^k$  where  $t$  is time. This does not hold when  $t \rightarrow 0$  (I have yet to meet the man who can draw a straight line exactly through minus infinity) nor when  $t$  is very large. If one turns a blind eye to these failings, it is possible to forget the basic empiricism and to believe the power law true, i.e. complete in itself and holding all ultimate meaning and explanation. The constants involved are elevated from convenient but meaningless numbers to the status of physical properties. Time no longer marches past in a linear and regular manner, but is distorted at the whim or fancy of a piece of rubber or a bit of cheese. "Mystical," which my dictionary defines as beyond human comprehension, is perhaps the most appropriate term.

Certain data in stress and strain may be accurately represented by a function containing  $\exp \sqrt{t}$ . This does not mean that the square root of time is real in physical fact; it arises from a certain, approximately normal, distribution of a physical property and in the mathematics of the Laplace transform required to translate the result of that distribution into observed behaviour. Fundamentally, and for every individual molecule, time is still behaving in the same old way. Failure to realize this aspect of mathematics and their application is largely responsible for the "mystical philosophical allusiveness" to which Dr. Lawrence so very rightly objects and which is so regrettably tending to taint certain aspects of rheology at the present time.

H. H. MACEY.

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EXETER.

### Mechanical models for the representation of visco-elastic properties

In his paper on "Mechanical models for the representation of visco-elastic properties" which appeared in your July issue, Dr. Roscoe expresses the opinion that the models  $(H|N)-N$  and  $(H-N)|N$  where  $H$  denotes an elastic spring,  $N$  a dashpot, the vertical bar parallel coupling, and the horizontal bar coupling in series, exhibit the same mechanical



behaviour. This statement does not apply in general. The point where Dr. Roscoe's arguments went wrong is where he put for the spring Force  $= n \times$  Extension and for the dashpot Force  $= \eta \times$  Rate of Extension and implicitly assumed that the latter kinematical quantity is the time-derivative of the former. To see that this will generally not be the case, consider a prism made of a viscous material, such as bitumen, to be extended by fixing one end at  $x = 0$  and moving the other end  $x = l$  with the velocity  $v$ . The velocity gradient in the direction  $x$  which is the kinematic variable governing viscous resistance, called by Dr. Roscoe "rate of extension," is  $dv/dx$  or assuming the deformation to be homogeneous  $= v/l$ , where  $v = dl/dt = \dot{l}$ . On the other hand if the material has also elasticity obeying Hooke's law the extension will be measured by  $e = \Delta l/l_0 = (l-l_0)/l_0$  where  $l_0$  is the original length. Now evidently the time derivative of  $e$  or  $\dot{e} = \dot{l}/l_0$  and this is different from  $v/l = \dot{l}/l$ . It is the same only when  $l$  differs so little from  $l_0$  that it can be replaced by the latter. There is a way out by defining the extension by the logarithmic measure, viz.  $e = \ln(l/l_0)$  in which case  $\dot{e} = \dot{l}/l$  whatever the magnitude of  $\Delta l$ . But it can be proved by tensor-analysis that his way out is only available in *pure* extension, i.e. in the absence of rotation of principal axes. In finite simple shear, for instance, there is no way of defining the rate of strain in such a manner that it is equal to the time-derivative of the strain. The mechanical models, however, which if actually built as shown could not be deformed by shear, have mostly been applied to infer "the behaviour of a material" in simple shear or in other deformations involving rotations of the principal axes. Therefore the argument of Dr. Roscoe is *in general not applicable* except in the very restricted cases when either (a) the strain of the spring is infinitesimal, or (b) when the deformation is *pure* and the strain is measured in the logarithmic measure. Both restrictions will be inapplicable just in those cases where the models are most useful. The rheological behaviour of Dr. Roscoe's models Fig. 5 (a) and (b) in the case of finite strain is the subject of a paper which will appear elsewhere.

M. REINER.

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Prof. Reiner apparently fails to realize that the writer's paper deals purely with equivalences between certain mechanical models which are frequently used for the repre-

sentation of visco-elastic properties. The models considered are built up of springs for which the extension is strictly proportional to the force, and dashpots for which the rate of extension is strictly proportional to the force; and it is to be noted that the rate of extension of a spring or dashpot means nothing more nor less than the first derivative of its extension with respect to time. Now it is sometimes found that the relation between a particular component of stress and the corresponding component of strain (defined in a suitable way) in a visco-elastic material is the same as the relation between the force and extension in a certain model; that is to say, the same linear differential equation which relates force and extension in a model is sometimes found to relate a component of stress and the corresponding component of strain in a material. The argument given in the paper is in no way concerned with the conditions under which such a formal correspondence may be found to exist, but deals only with the force-extension relationships for these models, so it is incorrect to suggest that the argument is "*in general not applicable*" except in the very restricted cases when either (a) the strain of the spring is infinitesimal, or (b) when the deformation is *pure* and the strain is measured in logarithmic measure." For, as regards (a) if the argument holds for infinitesimal extensions it necessarily holds for finite extensions since the models are linear. As regards (b) it must be noted that the term "strain" is not used in the paper in connexion with *models* so that the question of its measure is irrelevant.

Prof. Reiner's criticism seems to be based on a difficulty in defining the term "rate of strain," a difficulty which can only properly arise in considering *materials* since the term is superfluous in the case of models. In the brief reference to the properties of materials at the beginning of the paper, the writer has implied that a particular type of relation often exists between a particular component of stress, the corresponding component of strain in the material, and time; but the term "rate of strain" is nowhere used so that the criticism seems to be beside the point.

Finally, exception must be taken to Prof. Reiner's statement that the velocity gradient in a prism of viscous material is "called by Dr. Roscoe 'rate of extension.'" The writer has never called it by a name so obviously incorrect.

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## NOTES AND NEWS

## New Books

**Reports on Progress in Physics.** Vol. XIII (1950).  
(London: The Physical Society.) Pp. vi + 424.  
Price 50s. net.

This volume follows the pattern of previous volumes in selecting certain topics for review. By recording the progress made in each of them over a period of years, the several authors have in effect provided a collection of valuable monographs of varying length under one cover.

The first article on the "Investigation of Eye Movements," by Lord and Wright, is an example of the growing interest in the borderland between physics and physiology. Its main interest to applied physicists will probably be a discussion on involuntary eye movements which may affect estimates of position and distance.

"Infra-red Solar Spectroscopy," by Goldberg, deals mainly with a series of investigations carried out at the McMath-Hulbert Observatory at Michigan using Cashman lead sulphide photocells.

The article by Penney and Pike on "Shock Waves and the Propagation of Finite Pulses in Fluids" is a welcome account of a subject greatly extended during the war. It quotes freely from official reports now removed from restricted circulation. Indeed, 90 per cent of its references concern work carried out in the war years.

Professor Stoner's article on "Ferromagnetism: Magnetization Curves" is the longest in the volume. Experimental work on ferromagnetism is stimulated in no small degree by the technological requirements for materials with particular magnetic properties as deduced from magnetization curves, an astonishing variety of which have been recorded. This report is mainly concerned with the work of the past sixteen years, both experimental and theoretical, which has contributed to a better knowledge and understanding of the physical mechanism of ferromagnetic magnetization. There is much of interest in it to the applied physicist if he is prepared to dig. Perhaps special attention may be drawn to the interesting summary of the work of Shockley and others at the Bell Telephone Laboratories on the study of magnetic domains by coating the surface of single crystals with a colloidal suspension of magnetite.

Readers of the article on "Radio Astronomy," by Ryle, will note further examples of radio apparatus based on well-known optical devices. One of the latest is a spaced aerial interferometer analogous to the Michelson optical method of measuring stellar diameters, for the study of intense sources of radio waves in the galaxy.

Two articles then follow, one on "Planetary and Satellite Atmospheres," by Kniper, and another on "Paramagnetic Relaxation Effects," by Cook.

A well-written and illustrated account of "Cyclic Accelerators," by J. H. Fremlin and the late J. S. Gooden, includes useful constructional detail and compares the principles and capabilities of machines which use magnetic control of particle orbits.

The volume concludes with a long article on "Mesons," by C. F. Powell. The reviewer has watched the development of the photographic emulsion technique under Professor Powell from the days when very few nuclear physicists shared

his belief in its potentialities. In this lucid up-to-date account of the present state of the subject, information from all the standard techniques is gathered together and collated. Illustrated by 12 plates of nuclear events, it is an article written by an experimenter for experimenters.

A. M. TYNDALL

**Text Book of Physics, Part IV, Sound.** By J. DUNCAN and S. G. STARLING. (London: MacMillan and Co., Ltd.) Pp. xi + 105. Price 5s. net.

It is remarkable that *Duncan and Starling*, which has been reprinted frequently and held its place as a popular text-book of physics for science and engineering students for over thirty years, has, until now, remained in its first edition. In this second edition of the Sound section the original nine chapters on this subject are again reprinted, together with some eight or ten additional paragraphs and a like number of new exercises. Among the headings of these paragraphs are Noise, The bel and the decibel, The phon, Ultrasonic waves, Echo depth-sounding and The Gramophone, but, as is to be expected in a book of "Intermediate" standard, the treatment of these topics is an outline rather than a detailed one. Strangely the modern definition of *noise* seems to be omitted, but *Lissajou*, whom the reviewer seems to remember was content with a double *s*, appears, as is not unusual, in the plural. These are, however, trivial criticisms, and, even though this treatment of the elements of sound may be too classical or too thorough to be really fashionable, it should nevertheless continue to serve a useful purpose.

E. J. IRONS

**Some Applications of Nuclear Physics to Medicine.** By W. V. MAYNEORD, D.Sc., F.Inst.P. (London: The British Institute of Radiology.) Pp. xii + 290. Price 35s.

In February 1947 members of the British Institute of Radiology were privileged to hear a course of six lectures on applications of nuclear physics to medicine given by Professor Mayneord. Following his work in Canada in this field, these lectures were deservedly very popular among a mixed audience comprising medical radiologists, hospital physicists and engineers. The present work has been built around those original six lectures, although a considerable amount of extra material has been added.

As the author is at pains to emphasize, this Supplement is not intended as a text-book of the medical applications of nuclear energy; it is rather a general review of the subject highlighted with accounts in more detail of particular aspects with which the author and his colleagues have been particularly concerned.

The Supplement is divided into two parts: Part I commences with a general account of the physics of the nucleus and nuclear reactions. This section, which is almost identical with the text of the original lectures, is very readable although most medical radiologists will probably find it rather heavy going. The latter will no doubt find the inclusion of uranium 238 in a table of stable isotopes rather confusing. They will also look in vain under the heading "packing fraction" for



ny explanation of the term, but these are minor blemishes in an otherwise lucid summary.

Following the general physics is an account in some detail of the uses of five radioactive isotopes commonly employed in clinical techniques, namely phosphorus 32, iodine 131, iron 55 and 59, and sodium 24. Although the author admits that it is outside his province to assess the results of the medical uses of these isotopes, it is clear that, at present, their diagnostic applications are proving more important than their use in therapy. Phosphorus 32 and iodine 131, in particular, have largely disappointed the early optimism of some workers that these substances would prove powerful therapeutic tools against malignant disease.

In the diagnostic field, a fascinating account of many diverse uses is given. To mention but one example quoted by the author which is perhaps particularly ingenious—the use of iron 55 and 59 in the study of the effects of blood transfusion. The donor is given one form of iron and the recipient the other. In both cases some of the radio-iron is used to form “tagged” haemoglobin in the red cells. Observations made on the fate of the two types of radioactive cell have provided important information on the interaction of blood groups with the consequent safeguarding of transfusions.

For completeness, the author has included in Part 1 a section on health hazards arising from the use of radioisotopes. In addition to the Medical Research Council's *Introductory Manual on the Control of Health Hazards from Radioactive Materials*, there is included the Rules at present in force at the Royal Cancer Hospital, London. As a guide to others who may be faced with the problems attending the establishment of a radio-isotope service in a hospital, these rules seem admirable.

Part 2 of the Supplement is concerned with isotope dosimetry, with the addition of two extra sections on betatron and synchrotron measurements and on scintillation counting. The dosimetry section is likely to become the valued friend of every hospital physicist. The large amount of information which has been packed into the compass of fifty pages is truly incredible, particularly when it is realized that in almost all cases are the proofs of necessary mathematical formulae set out in full. It is true that much of this section is not original and has appeared scattered throughout the literature, but the author has rendered an inestimable service in presenting in one place such an admirable summary. Incidentally, he has dealt at some length in this section on the physics of cobalt 60 as a gamma-ray source suitable for replacing radium both in needle form and in “telecurie” units. The advantages of cobalt 60 over radium in the latter type of unit, both in the increased gamma-ray flux and the greater depth dose made possible by using larger sources at greater distances from the patient, are obvious, but the protection problems attendant on the use of sources of possibly 100 or even 1 000 curies are formidable.

So much has been written recently on the acceleration of atomic particles that it is doubtful whether the few pages devoted to a rather sketchy account of the action and potentialities of various accelerators are worthwhile. However, this leads naturally to the section relating some measurements on radiation beams from betatrons and synchrotrons both in America and in this country made by the author and one of his colleagues, Dr. J. H. Martin.

Rather unexpectedly, a section on scintillation counting

completes the Supplement. An experimental arrangement is described using a calcium tungstate crystal in conjunction with an R.C.A. IP21 photomultiplier, which has been used for various investigations. Some clinical applications of the method are described and a brief comparison made with results obtained with the more conventional Geiger-Müller tubes.

This Supplement, as well as proving a valuable addition to the library of the hospital physicist, will serve as a good introduction to the subject for scientific workers in allied fields. The bibliography is excellent throughout. Although the table of contents is adequate, the absence of an index is disappointing in a work so abundant with references. It is also to be regretted that no exception was made with regard to the paper cover used in this series of Supplements. In a work destined to be so well thumbed, a stiff binding would have considerably enhanced the book's value. Nevertheless, in other respects the Supplement is well produced. The paper is good and the diagrams are clearly reproduced and the book is well worth its modest price.

W. A. LANGMEAD

### Co-operative Industrial Research—A study of the economic aspects of the Research Associations grant-aided by the Department of Scientific and Industrial Research.

By R. S. EDWARDS. (London: Sir Isaac Pitman and Sons Ltd.) Pp. xiv + 285. Price 20s. net.

This is a most valuable book, packed with information and readable. The author has obviously written every word of it himself, and with enthusiasm at that. He must also have written or approved the statement on the dust jacket

“This book will fill a gap in the literature of industrial organization. It appraises one of the most interesting developments of the last thirty years—co-operative industrial research financed jointly by industry and the government. The book falls into four parts: Part I deals with the theory of co-operative research and the types of institution through which it can be arranged; Part II traces the development of the research association scheme; Part III discusses current problems of finance, including the contentious subject of compulsory levies; Part IV reviews the functions of research associations and assesses their potentialities for the future. Information and views were collected from over two hundred firms and these are summarized and examined. The object of the study is twofold: first, to provide material for those concerned with the future of research, and, second, to throw light on certain general questions of industrial structure which have become important in recent years.”

which, remarkable though it might seem to a cynic, is nothing but the truth.

All through the book the shrewd, pleasant personality of Professor Edwards keeps peeping out. He makes good observations of every kind; for example: “Another point which must not be overlooked is that the majority of firms do not in fact aspire, and may be wise not to aspire, to scientific and technical leadership.” There is a very good index—try “O” for “Operational Research.” It is sincerely to be hoped that this book will not get lost in the great crowd of present-day technical publications. Finally, it is pleasant to note that during the time the book was in the press, the author was made a member of the Advisory Council for Scientific and Industrial Research.

F. A. FREETH

## Notes and Comments

### Elections to The Institute of Physics

The following elections have been made by the Board of The Institute of Physics:

**Fellows:** J. M. Buist, J. A. Clegg, S. M. Cox, M. R. Gridley, E. H. Jones, R. S. T. Kingston, M. P. Lord, G. E. Roth, J. W. Sharpe.

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Seventy graduates, thirty subscribers and ninety-three students were also elected.

**Certificates in Laboratory Arts.**—Twenty-five candidates from two colleges entered for one or more sections of the Laboratory Arts examination and ten candidates successfully completed the necessary three sections to qualify for certificates. The sections, of which section A is compulsory, are: A. Physics and Mathematics. B. Simple Wood and Metal Work. C. Glass-blowing. D. Laboratory Organization and Technique.

**National Certificates in Applied Physics.**—For the 1950 examinations for National Certificates in Applied Physics there was again a substantial increase in the numbers of candidates. The existence of these certificates, for which the first examination was held as recently as 1947 (when one college entered candidates), is becoming widely known; twelve colleges presented seventy candidates for the Ordinary Certificate and one college (Northampton Polytechnic, London) presented the first six candidates yet to enter for the Higher Certificate. There were forty-four successful candidates at the Ordinary stage and four at the Higher.

**Journal of Atmospheric and Terrestrial Physics.**—We have received a copy of the first number of a new periodical—*The Journal of Atmospheric and Terrestrial Physics*. The Editor-in-chief is Sir Edward Appleton, and there is a strong international editorial advisory Board.

In his foreword Sir Edward writes: "... it cannot be denied that, because of the increasing volume of scientific papers and because of the greater specialization of much of our modern scientific activity, there is a real need to supplement these (learned society publications) general channels of publication by others designated to cater for more limited fields of scientific interest. This *Journal*, then, may be counted such a specialized channel of scientific publication, although it will be by no means our policy to restrict the ambit of its subject matter too narrowly." In addition to scientific papers of normal length it is proposed to publish shorter communications under the heading of Research Notes.

Brief notices describing geophysical events of special interest will also be welcomed. The acceptable languages are English, French and German.

The periodical is to appear at first bi-monthly, but may appear very frequently later on. The subscription price for the volume of about 300 pages is £3 10s. 0d., and it is published by Butterworth-Springer Ltd., of London.

## Journal of Scientific Instruments

### Contents of the December issue

#### ORIGINAL CONTRIBUTIONS

Geiger-counter X-ray spectrometer—influence of size and absorption coefficient of specimen on position and shape of powder diffraction maxima. By A. J. C. Wilson.

Orientation of crystals using a Geiger-counter X-ray spectrometer. By J. A. James.

A note on the calibration and use of a shielded hot-wire anemometer for very low speeds. By C. F. Cowdrey.

Some mechanical features of a double-crystal neutron spectrometer. By G. E. Bacon, J. A. G. Smith and C. D. Whitehead.

A method for the adjustment of the compensator plate of a Michelson interferometer. By J. E. H. Brayton.

Apparatus for the study by electron diffraction of the effect of temperature on surface films. By J. W. Menton and J. V. Sanders.

A low-temperature microscope stage. By R. G. Rhodes.

#### NOTES AND NEWS

##### Correspondence.

Focusing in electron microscopy. By R. S. M. Revell and A. W. Agar.

A demountable accelerator tube. By D. R. Chick and F. J. Miranda.

The setting of single crystals from zero layer curve photographs. By P. T. Davies.

##### New Instruments, Materials and Tools.

Pneumatic infra-red detector—Ten-channel strain gauge equipment—Artificial single crystals of optical quality—Permanent magnet electron microscope—Sub-miniature electrometer triode—Gamma-ray detector—Stainless steel Geiger-Müller tubes—High voltage X-ray therapy set—Glass electrode for use with alkaline solutions.

##### New Books.

The Magnetic Amplifier—An Introduction to Electronics—Techniques Generales du Laboratoire de Physique—Testing Radio Sets—Micro-wave Measurements—Industrial Instrumentation—The Binocular Microscope.

##### Notes and Comments.

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